Development of innovative landfill gas management technologies

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This PhD project followed the Industrial PhD Programme offered by Innovation Fund Denmark (Innovationsfonden) and was conducted as a collaboration between The Technical University of Denmark (DTU) and NIRAS A/S.

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Preface

The work presented in this PhD thesis was carried out as an industrial PhD project in collaboration between NIRAS A/S and Department of Environmental Engineering at the Technical University of Denmark (DTU). Supervisors at the university were Professor Peter Kjeldsen and co-supervisor Professor Charlotte Scheutz. Supervisors from NIRAS were Anders G. Christensen and co-supervisor Jens E. Larsen. The project was carried out from October 2014 to January 2019 (including maternity leave and two three-month (six months in total) periods of leave to work on other projects). The PhD project was funded mainly by Innovation Fund Denmark (Project ref. no.: 4135-00011B) with additional contributions from DTU, NIRAS A/S and ARGO I/S.

The thesis is organised in two parts: the first part puts into context the findings of the PhD in an introductive review, and the second part consists of the papers listed below. These will be referred to in the text by their paper number, written with the Roman numerals **I-III**.

I Fjelsted, L., Christensen, A.G., Larsen, J.E., Kjeldsen, P., Scheutz, C., 2018. Assessment of a landfill methane emission screening method using an unmanned aerial vehicle mounted thermal infrared camera – A field study. *Waste Management* - in press. [Paper **I]**
doi:10.1016/j.wasman.2018.05.031

II Fjelsted, L., Christensen, A.G., Larsen, J.E., Kjeldsen, P., Scheutz, C., 2018. Closing the methane mass balance for an old closed Danish landfill. Accepted (with revision) by *Waste Management*. [Paper **II**]

III Fjelsted, L., Scheutz, C., Christensen, A.G., Larsen, J.E., Kjeldsen, P., 2018. Biofiltration of diluted landfill gas in an active loaded open bed compost filter. Submitted to *Waste Management*. [Paper **III**]

In addition, the following publications, not included in this thesis, were also concluded during this PhD study:

Fjelsted, L., Thomasen, T.B., Valbjørn, I.L., Scheutz, C., Christensen, A.G., Kjeldsen, P., 2015. Development of an innovative UAV-mounted screening tool for landfill gas emissions. *Proceedings Sardinia 2015, Fifteenth International Waste Management and Landfill Symposium,* S. Margherita di Pula, Cagliari, Italy.

Fjelsted, L., Scheutz, C., Christensen, A.G., Kjeldsen, P., 2017. Screening tool for landfill gas emission hotspots based on infrared images. *Proceedings Sardinia 2017, Sixteenth International Waste Management and Landfill Symposium,* S. Margherita di Pula, Cagliari, Italy.

In this online version of the thesis, papers **I-III** are not included but can be obtained from electronic article databases, e.g. via www.orbit.dtu.dk, or on request from DTU Environment, Technical University of Denmark, Bygningstorvet, Building 115, 2800 Kgs. Lyngby, Denmark, info@env.dtu.dk

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I also wish to thank *Finn Kjær* at ARGO I/S for giving me access to use their landfills in my research and for the support provided during the project.

As an industrial Ph.D. I had to cope with two workplaces, two desks and two sets of stakeholders, but I also had to double up on great colleagues and double up on skilled persons willing to help where my abilities were not sufficient – I am deeply grateful for all your support. From DTU Environment special thanks go to:

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Summary

Climate change is on the political agenda worldwide, and abatement strategies for greenhouse gas emissions are a necessity. One source of greenhouse gas emissions is landfills, as the degradation of organic carbon in landfilled waste generates methane. Landfill gas emission patterns show high spatial and temporal variability, but the development of innovative technologies for both monitoring and mitigation will help in the much-needed conceptual understanding of governing gas transport and emission processes.

A methane mass balance can be established based on the individual migration pathways for the generated methane, including methane recovery for energy utilisation or flaring, lateral migration to neighbouring plots, methane oxidation by microorganisms in the cover and emissions seeping into the atmosphere. A methane mass balance forms a good conceptual framework for setting up a mitigation strategy for a landfill.

The main part of this PhD project was conducted in relation to a closed Danish landfill (Hedeland landfill, Roskilde, Denmark). Many years of intensive investigations have been conducted at Hedeland landfill to understand better the migration and emission patterns of methane generated at the site. A mitigation strategy has to be established, which takes into consideration both the safety of local residents and the negative impact on global warming from landfill gas migration and emission. A methane mass balance for the landfill could provide a valuable overview and show the individual importance of each migration pathway.

As part of this PhD project, a methane mass balance was established for Hedeland landfill based on data from many years of investigation, covering the years 2013-2015. Methane generation was modelled based on a multi-phase, first-order degradation kinetics (Afvalzorg) model, with average methane generation determined at 67±8.6 kg h^{-1}. Methane recovery, emission and lateral migration were found to cover 38% of the modelled methane generation, each accounting for an equal share (9 ±2.9, 8 ±4.1 and 9 ±2.4 kg h^{-1}, respectively). Methane oxidation in the cover was identified as the migration pathway capable of closing the mass balance and accounting for the remaining 62% of the generated methane. Several indications supported a high oxidation rate in the landfill cover at Hedeland, including a low total emission rate, which was determined using the tracer gas dispersion method and a few emission hotspots with elevated methane concentrations at the

surface (identified by screening the whole landfill surface, using a flame ionisation detector).

Identification of landfill gas emission hotspots is the basis for establishing emission abatement technologies such as biocovers. To overcome the high spatial and temporal variability of landfill gas emissions, a screening tool based on an unmanned aerial system mounted with a thermal infrared (TIR) camera was tested at two Danish landfills (Hedeland and Audebo landfills). The correlation between landfill gas emissions (methane and carbon dioxide), surface temperatures obtained with the TIR camera and soil temperatures at 5- and 10-cm depths was investigated in an established test area at each of the two sites. At Hedeland landfill, no correlation was found between gas emissions and surface temperatures. In addition, identified methane surface fluxes were very limited, with an average for the four measuring campaigns of only 1.3 ±16 g CH_4 m^{-2} d^{-1}. An average methane flux of 371 ±1337 g CH_4 m^{-2} d^{-1} was found at Audebo landfill for five measuring campaigns. Furthermore, elevated temperatures at both the surface and at 5- and 10-cm depths were found in the same area as where the highest landfill gas surface fluxes were measured, thus indicating that in the right conditions the TIR camera could be used for delineating landfill gas emissions. A minimum flux of 150 g CH_4 m^{-2} d^{-1} from an area of at least 1 m^2 was established as the limit for the TIR camera being able to delineate a landfill gas emission hotspot at a typical Danish landfill.

When landfill gas is mixed with air it dilutes, often with a methane content too low for utilisation. However, mitigation is still needed to minimise the negative impacts on the environment, and to ensure human health and safety. Sources to dilute landfill gas could be remediation systems for lateral migration, emissions from leachate and monitoring wells or from air penetrating the cover of the landfill. A cost-efficient mitigation technology for dilute landfill gas could be microbial oxidation in an actively loaded biofilter. This technology was tested in an open-bed pilot-scale compost filter at Hedeland landfill, constructed in a 30 m^3 container. The filter was loaded with landfill gas diluted with ambient air to a methane concentration of between 5 and 10 vol.%. The filter was tested in five flow campaigns with the same methane inlet concentration and an increasing methane load between 106 and 794 g CH_4 m^{-2} d^{-1}. The highest observed methane oxidation rate was 460 g CH_4 m^{-2} d^{-1} with an oxidation efficiency of 58%. Overall, oxidation efficiencies of more than 87% were never achieved, due to substantial preferential flows at the transition point between the compost and container

wall despite an attempt to design the container with blockers against preferential flows. However, pore gas profiles showed methane oxidation of 100% in the compost material. These results were supported by tracer gas tests showing an average methane oxidation of almost 86% at 10 cm below the surface of the filter in flow campaign 5, where the load had an average of 701 ±47 g CH_4 m^{-2} d^{-1}.

At Hedeland landfill, three remediation systems have been installed to cut off laterally migrating landfill gas from reaching residential houses on neighbouring plots. In 2017, an average methane content of 0.53 ±0.55 vol.% in off-gas from these remediation systems was observed, accompanied by an oxygen content in most cases above 10 vol.%. Treatment of the remediation off-gas in the constructed pilot-scale biofilter would result in a methane load of 717 g CH_4 m^{-2} d^{-1}. Nevertheless, the gas retention time would only be 3 min, due to the high pump flow rate of 80 m^3 h^{-1}, which is thought to be below a critical gas retention time. To increase the retention time to 30 min, ten containers similar to the tested filter would be needed. A suggested alternative could be a 111 m^2 biofilter embedded in the landfill cover, which would result in the same load as the ten containers. An embedded biofilter is also expected to be able to overcome the challenges of preferential flows experienced in the tested container solution.

Dansk sammenfatning

Verden over er de globale klimaforandringer på den politiske dagsorden, og der er et stigende behov for strategier, som kan mindske udledningen af drivhusgasser. En kilde til udledningen af drivhusgasser er deponeret affald, hvor nedbrydningen af organisk kulstof danner metan. Emissionen af deponigas er meget dynamisk med en stor rumlig og tidslig variation. Der er derfor behov for udvikling af innovative teknologier til både måling og håndtering af deponigas, som kan hjælpe i den nødvendige konceptuelle forståelse af, hvad der styrer gastransporten og udledningsprocesserne.

En metanbalance kan opstilles på baggrund af den dannede metans forskellige migrations veje, som inkluderer opsamling med henblik på energiudnyttelse eller afbrænding, horisontalt udslip til naboejendomme, metanoxidation i dæklaget samt emission til atmosfæren. En god konceptuel ramme for udarbejdelsen af en håndteringsstrategi for et deponi er en metan massebalance.

Størstedelen af nærværende Ph.d. projekt blev udført i forbindelse med et nedlukket dansk deponi (Hedeland deponi, Roskilde). Mange års intensive undersøgelser har været gennemført på Hedeland deponi, for bedre at forstå under hvilke forhold den dannede deponigas spredes og emitteres til atmosfæren. En håndteringsstrategi, der tager højde for både sikkerheden for naboerne og de negative effekter på klimaet, er nødvendig. En metan massebalance for deponiet kan give et overblik og vise betydningen af de enkelte migrations veje i forhold til hinanden.

Som en del af dette Ph.d. projekt er der opstillet en metan massebalance for Hedeland deponi baseret på de store mængder data, der er indsamlet i forbindelse med de mange års undersøgelser, og som dækker årene 2013-2015. Metandannelsen blev modelleret ved brug af en multifase-model baseret på førsteordens nedbrydningshastigheder (Afvalzorg) og den gennemsnitlige metandannelse blev fundet til 67 ±8.6 kg h^{-1}. Metanindvindingen, emissionen og den horisontale migration viste sig at udgøre 38% af den modellerede dannede metan, og de bidrog med en lige stor andel hver (hhv. 9 ±2,9, 8 ±4,1 and 9 ±2,4 kg h^{-1}). Metanoxidation i dæklaget blev identificeret som den spredningsvej, der kunne lukke metanbalancen, og den udgjorde 62% af den dannede metan. Der var adskillige indikationer, som støttede en høj oxidationsrate i dæklaget på Hedeland deponi herunder en lav total emissionsrate bestemt ved brug af sporgasmetoden og få emissions-hotspots med forhøjede koncen-

trationer af metan på overfladen (fundet ved en overflade screening med en flammeioniserings detektor).

Identifikation af emissions-hotspots med lossepladsgas er grundlaget for etableringen af emissionsbegrænsende teknologier så som biocovers. En drone udstyret med at termokamera blev testet på to danske deponier (Hedeland deponi og Audebo deponi) som et muligt screeningsværktøj, der vil kunne imødegå den store rumlige og tidslige variation i lossepladsgas emissionen. Sammenhængen mellem udledningen af lossepladsgas (metan og kuldioxid), overfladetemperaturen målt med termokameraet og jordtemperaturen i 5 og 10 cm dybde blev undersøgt i et testområde på hver er de to deponier. På Hedeland deponi blev der ikke fundet nogen sammenhæng mellem emission af deponigas og overfladetemperaturene. Desuden var de fundne overfladefluxe meget begrænsede, hvor gennemsnitsfluxen for de fire målekampagner var begrænset til 1,3 ±16 g CH_4 m^{-2} d^{-1}. En gennemsnitlig metanflux på 371 ±1337 g CH_4 m^{-2} d^{-1} blev fundet for Audebo deponi for de fem gennemførte målekampagner. Derudover blev der fundet højere temperaturer både på overfladen og i 5 og 10 cm dybde i de samme områder, hvor de højeste overfladefluxe af deponigas var målt, hvilket indikerer, at termokameraet vil kunne være i stand til at identificere emissioner af deponigas under de rette forhold. En minimumsflux på 150 g CH_4 m^{-2} d^{-1} fra et område på mindst 1 m^2 blev fundet som den nedre grænse for hvornår et termokamera er i stand til at identificere et emissions-hotspot med deponigas på et typisk dansk deponi.

En fortynding af deponigas med luft vil ofte resulterer i et metanindhold i den fortyndede deponigas som er for lavt til at gassen kan udnyttes. Dog vil der ofte stadig være behov for at minimere de negative effekter på miljøet og for menneskers sikkerhed. Kilderne til fortyndet deponigas kan være afværgesystemer imod den horisontale spredning af deponigas, emissioner fra perkolat- og overvågningsbrønde eller ved at luft trænger ned i dæklaget på deponiet. En kosteffektiv begrænsningsteknologi for fortyndet deponigas kan være mikrobiel oxidation i et biofilter med aktiv tilførsel af gas. Denne teknologi blev testet i et kompostbaseret åbent pilotskala filter på Hedeland deponi konstrueret i en 30 m^3 container. Filteret blev tilført deponigas fortyndet med luft til en metankoncentration på mellem 5 og 10 vol.%. Filteret blev testet i fem flowkampagner med den samme metankoncentration i indløbet i alle fem kampagner, men med en stigene metantilførsel, der resulterede i en tilførsel på mellem 106 og 794 g CH_4 m^{-2} d^{-1}. Den højeste fundene metanoxidationsrate var 460 g CH_4 m^{-2} d^{-1} med en oxidationseffektivitet på 58%. Den højeste samlede oxidationseffektivitet, der

blev fundet var 87%, og en højere effektivitet blev aldrig opnået på grund af væsentlig præferentiel transport langs siderne mellem komposten og containervæggen til trods for de installerede blokader designet til at begrænse det selektive flow. Dog viste gasprofiler i komposten en metanoxidation på næsten 100%. Test med en sporgas understøttede disse resultater og viste en metanoxidation på 86% 10 cm under filteroverfladen ved flowkampagne 5, hvor der gennemsnitligt blev tilført 701 ±47 g CH_4 m^{-2} d^{-1} til filteret.

På Hedeland deponi er der installeret tre afværgesystemer for at afskære den horisontale spredning af lossepladsgas til beboelsesejendommene på nabogrundene. I 2017 blev der målt et gennemsnitlig metanindhold i den oppumpede gas fra alle tre anlæg på 0,53 ±0,55 vol.% og med et indhold af ilt på over 10 vol.% i de fleste tilfælde. Håndteringen af den oppumpede afværgegas i det testede pilotskala biofilter vil resultere i en metan tilførsel på 717 g CH_4 m^{-2} d^{-1}. På grund af den høje pumperate på 80 m^3 h^{-1} vil gassens opholdstid i filteret dog kun være på 3 minutter, hvilket forventes at være mindre end den nødvendige opholdstid. Ti filtre med det samme volumen som det testede filter er nødvendigt for at øge opholdstiden til 30 min. Et foreslået alternativ kunne være et 111 m^2 biofilter indbygget i deponiets slutafdækning, som vil resultere i den samme mængde metan tilført pr. arealenhed som til de ti containere. Et biofilter indbygget i dæklaget kan også være en løsning på problemerne med det selektive flow ved containerløsningen.

Table of contents

Abbreviations

CH_4	Methane
CO_2	Carbon dioxide
FID	Flame ionisation detector
GDL	Gas distribution layer
GHG	Greenhouse gas
HMPGS	Horizontal multi-port gas sampler
LFG	Landfill gas
MSW	Municipal solid waste
O_2	Oxygen
TIR	Thermal infrared
UAS	Unmanned aerial system
UAV	Unmanned aerial vehicle
VWC	Volumetric water content

1 Introduction

1.1 Background

Climate change is a global concern with a high political focus. One important greenhouse gas (GHG) is methane (CH_4) with a global warming potential 28 times that of carbon dioxide (CO_2) over a 100-year period, not taking into account climate feedbacks (IPCC, 2013). One of the causes of CH_4 in the atmosphere is the emission of landfill gas (LFG) with typical concentrations of CH_4 at 55-60 vol.% and 40-45 vol.% of CO_2 (Bogner et al., 2008). CH_4 emissions from landfills and wastewater treatment plants accounted for 18% of global anthropogenic CH_4 emissions in 2004-2005, or about 90% of total waste sector emissions (corresponding to about 3% of total anthropogenic GHG emissions) (Bogner et al., 2008). GHG emissions from landfills account for 49% of total GHG emissions from the Danish waste sector (Nielsen et al., 2018). LFG migration and emissions not only have an impact on the global greenhouse effect, but they also pose a risk to human health and safety, as CH_4 can be explosive when mixed with air at certain ratios in confined spaces (Christophersen and Kjeldsen, 2001; David J.V. Campbell, 1996; Franzidis et al., 2008; Williams and Aitkenhead, 1991).

The mitigation of LFG emissions covers different technologies, such as gas collection and recovery for energy utilisation (Stegmann, 1996; Willumsen and Barlaz, 2011) or thermal conversion of CH_4 to CO_2 through biological treatment in passive biocover systems or in actively loaded biofilters (Bogner et al., 2008). What solution transpires to be the most efficient in terms of both mitigation efficiency and cost efficiency depends on the specific site conditions and is influenced by the landfill design, the type and amounts of waste, the age of the landfill, physical installations and how LFG migrates (Aghdam et al., 2018a; Bogner and Spokas, 1993; Börjesson et al., 2009; Scheutz et al., 2011b; Spokas et al., 2006).

Mitigating the negative effects of LFG on the environment and on human health and safety requires site-specific knowledge about LFG migration pathways, and how significant they are at a given landfill can be illustrated through a CH_4 mass balance, which could form the basis for an effective LFG management strategy. LFG can be utilised for energy production, and so some landfills have installed a gas recovery system. The rest of the generated CH_4 can migrate either laterally to neighbouring plots or to the top cover of the landfill, where a certain quantity will be oxidised by methanotrophs in the

cover soil and the rest will be emitted into the atmosphere. A CH_4 mass balance for a landfill can be summarised as follows (Bogner and Spokas, 1993):

CH_4 generated = CH_4 recovered + CH_4 emitted + CH_4 oxidised + CH_4 migrated + ΔCH_4 storage

CH_4 generation is often modelled based on the specific waste fractions and amounts in models such as Afvalzorg, LandGEM or IPCC (IPCC, 2006; Jacobs and Scharff, 2001; Mønster et al., 2011; Mou et al., 2015; Scharff et al., 2000; Scharff and Jacobs, 2006). CH_4 recovery is often the most well-known pathway and can be calculated based on data from the collection system. Laterally migrating LFG is often calculated in the guise of diffusion through the bottom and side liners, under the assumption that the liners have not been compromised (Spokas et al., 2006). Changes in CH_4 storage are a function of many parameters (e.g. CH_4 concentration, void space, temperature and atmospheric pressure) and are probably the most difficult issue to quantify (Spokas et al., 2006). CH_4 oxidation in the landfill cover can be determined using different techniques, for example point measurements of soil gas profiles in the cover (e.g. Gebert et al., 2011; Röwer et al., 2011; Scheutz et al., 2011b), a carbon mass balance comparing raw LFG carbon content with carbon flux measurements at the surface (e.g. Christophersen et al., 2001; Scheutz et al., 2011a) or using the fractionation of stable carbon isotopes (e.g. Börjesson et al., 2007; Chanton et al., 2011, 2008, 1999; Scheutz et al., 2009). Different techniques can also be used to measure landfill CH_4 emissions, based on surface point measurements using a static or dynamic flux chamber, from which the total emission is interpolated (e.g. Mønster et al., 2019; Scheutz et al., 2008, 2003), or on remote sensing systems such as the tracer gas dispersion method, LiDAR or eddy covariance, which measures emissions from larger areas or across the whole landfill site (e.g. Babilotte et al., 2010; Mønster, 2014; Mønster et al., 2019, 2015, 2014; Rees-White et al., 2018; Scheutz et al., 2011). A few CH_4 mass balances for a whole landfill site have been reported in the literature (Aghdam et al., 2018; Bogner and Spokas, 1993; Börjesson et al., 2009; Scheutz et al., 2011b; Spokas et al., 2006), which could help understand the fate and migration pathways of LFG.

Sources of surface CH_4 emissions can be installations in the landfill, such as leachate wells, or perhaps cracks or weaknesses in the cover (emission hotspots) that can result in visible or more diffuse surface emissions. Identifying CH_4 emission hotspots is necessary in order to install an efficient abatement system. Installations such as leachate wells and monitoring wells in the

landfill are often easy to identify, and CH_4 emissions from these points can be quantified, for example, by using a tracer gas method (e.g. Fredenslund et al., 2010). The more diffuse CH_4 emissions from the surface of the landfill are more difficult to identify and delineate, as CH_4 emissions tend to vary significantly, both temporarily and spatially (Röwer et al., 2011; Scheutz et al., 2014; Xu et al., 2014). Furthermore, the size of an emission hotspot can be as low as just a few square centimetres, thereby making them more difficult to identify. Remote sensing systems are able to identify sizeable emissions from larger areas, but these methods are often not able to locate precisely the point of emission, which instead requires more detailed investigations that are often conducted nowadays using ground-based screening tools based on hand-held CH_4 sensors such as flame ionisation detectors (FIDs) or leak detection sensors for natural gas distribution systems (e.g. LaserOne from Huberg, Italy). These ground-based screening tools are time-consuming and not cost-efficient. Combining the advantages of remote sensing with the advantages of very detailed mapping, by using an unmanned aerial system or vehicle-mounted (UAS/UAV) sensor, has been investigated in a few studies (Allen et al., 2018, 2015; Berman et al., 2012; Capodici et al., 2015; Khan et al., 2012; Lando et al., 2017; Manzo et al., 2017; Nathan et al., 2015; Tanda et al., 2017). One type of sensor that has been tested, which today has a size and weight suitable for a UAV and still has a high resolution, is the thermal infrared (TIR) camera (Battaglini et al., 2013; Capodici et al., 2015; Desideri et al., 2007; Lewis et al., 2003; Madruga et al., 2007; Raco et al., 2005; Tanda et al., 2017). Anaerobic degradation and the generation of CH_4 are exothermic processes, and so LFG has a significantly higher temperature than surrounding areas (Hanson et al., 2010; Yoshida et al., 1999) and LFG migrating to the surface of the landfill could potentially leave a heat print at the surface of the landfill cover. These warmer areas could possibly be observed with a TIR camera. The few studies that have been conducted using a TIR camera for detecting LFG emission hotspots have resulted in various findings, and so further research is needed to investigate the UAV-mounted TIR camera's ability to delineate LFG emission hotspots.

Landfill CH_4 generation declines over time, thereby decreasing the amount of CH_4 in LFG as well as the potential for utilising generated LFG. Ultimately, this results in diluted LFG when ambient air starts to penetrate the cover. Other ways of diluting LFG could be from leachate wells or other installations in the landfill, where ambient air could mix with the LFG (Cassini et al., 2017; Scheutz et al., 2017), or another option could be off-gas from remediation sys-

tems installed to prevent LFG from migrating to neighbouring plots. The content of CH_4 in the diluted gas is often too low for any utilisation or flaring, but emissions into the atmosphere still have a potential impact on the climate. A cost-efficient treatment solution in this regard could involve microbial oxidation in a biofilter.

The main part of the research conducted on the biological treatment of LFG has concentrated on traditional compost-based filters treating pure LFG consisting mainly of CH_4, CO_2 and some trace gases or, in the case of laboratory column tests, pure CH_4 (e.g. Kjeldsen and Scheutz, 2019; Scheutz et al., 2009). In these cases, O_2 diffuses into the filter from the top and CH_4 or LFG is loaded from the bottom of the filter. A few studies have been conducted on the biological treatment of dilute LFG, where O_2 is actively loaded into the filter (Cassini et al., 2017; Farrokhzadeh et al., 2017; Haththotuwa et al., 2012; Haubrichs and Widmann, 2006; Park et al., 2009; Scheutz et al., 2017; Streese and Stegman, 2003; Thomasen et al., 2019). The main elements of these studies were conducted as laboratory tests, with only the studies by Cassini et al. (2017), Scheutz et al. (2017) and Streese and Stegmann (2003) conducted in the field. As laboratory column tests are conducted in stable and controlled conditions, field tests are needed to determine how the technologies perform at a landfill site with variable conditions and changing seasons.

1.2 Research objective

The overall objective of this study was to develop strategies, methodologies and technologies for managing gas-related environmental impacts at landfills. The specific objectives of the study were to:

- Establish a CH_4 mass balance model to evaluate the mitigation strategy of an old landfill by measuring CH_4 emission rates, estimating lateral CH_4 migration rates, collecting data for CH_4 recovery rates, measuring CH_4 oxidation rates and modelling CH_4 gas generation rates.
- Develop and demonstrate the use of a UAV-mounted thermal infrared camera as a landfill gas emission hotspot screening tool. In addition, investigate how surface soil temperatures and LFG emissions are correlated.
- Determine the CH_4 oxidation capacity of dilute landfill gas containing oxygen in an actively loaded, compost-based pilot-scale biofilter.

2 Materials and methods

2.1 Landfill site descriptions

The main part of the investigation in this PhD project was conducted at a closed Danish landfill, namely Hedeland landfill near Roskilde, which was established in 1979 in a former gravel pit. Approximately 2.9 million tonnes of mainly non-combustible waste and soil was landfilled there until operations were terminated in 2009. A cross-section through Hedeland landfill can be found in Figure 1, illustrating how it was constructed with a polymer liner on the bottom and the lower parts of the side slopes, while the upper part of the side slopes consisted of a clay liner. Leachate was drained from the whole landfill into the same leachate well. Gas recovery systems were installed in two sections of the landfill but did not cover the total site. The recovered gas was utilised in a dual-fuel engine producing electricity, with diesel oil as a support fuel. The landfill top cover consists of at least one metre of soil with no polymer liner, and seven residential houses are situated within a radius of fewer than 100 metres from the edges of the landfill. Many years of investigation in the area have uncovered laterally migrating landfill gas at a level posing a risk of explosion for five of the seven houses. To protect the residents in the five houses, three remediation systems have been installed, pumping the migrating landfill gas from some of the monitoring wells installed in the soil compartment adjacent to the site. For a more detailed description of the construction of the landfill, waste categories, annual landfilled waste amounts, the gas collection system and the laterally migrating gas remediation system, see papers **I** and **II**.

Part of this investigation was also conducted at Audebo landfill, located close to Holbæk, Denmark, and owned by the same waste utility company as Hedeland landfill. Audebo landfill was established in 1990 and is still in operation. Three sections of the landfill have a permanent top cover, and these contain approximately 604,000 tonnes of mainly non-combustible waste, albeit in the first section some municipal solid waste has been landfilled. Landfilling in these three sections was terminated in 2009. The landfill was established with leachate collection and was covered with one metre of soil on top of a root-blocking material. More details are available in paper **I**.

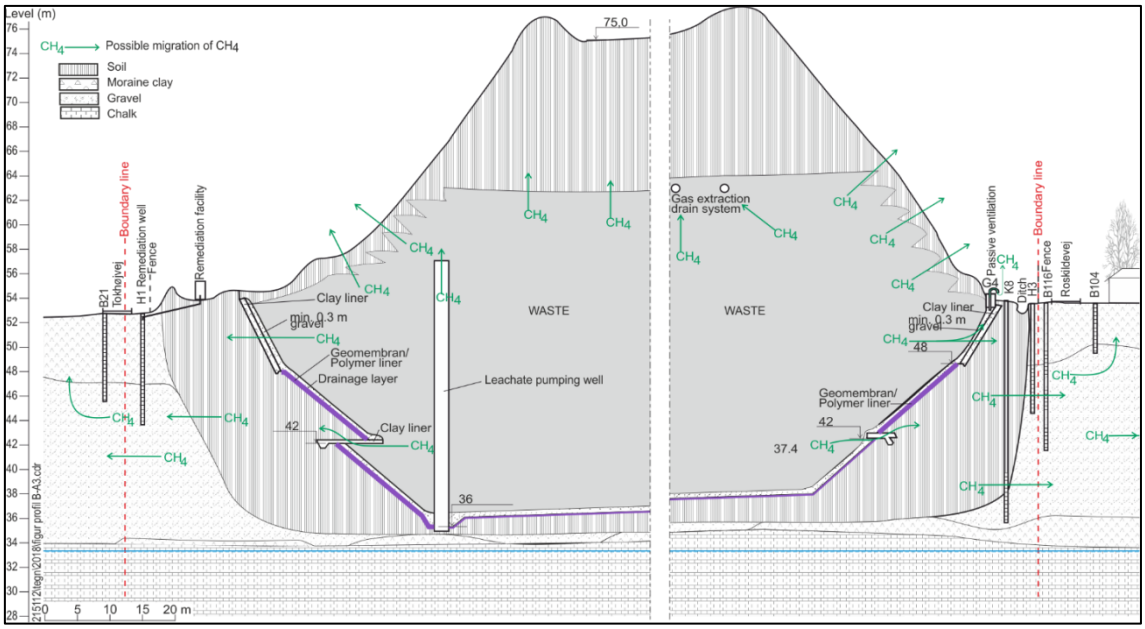

Figure 1. Cross-section of Hedeland landfill. The middle of the figure has been removed. Note that the scale is not the same in the vertical and lateral directions. From paper II.

2.2 Landfill methane mass balance

Numerous measurements have been conducted through the years at Hedeland landfill, in order to understand the migration of LFG from the site, both vertically into the atmosphere and laterally toward neighbouring plots. These data were collected and analysed together with data about landfilled waste amounts and waste fractions, as well as from the gas recovery system. From these data a CH_4 mass balance was established in this study for Hedeland landfill covering the years 2013-2015.

Methane generation was modelled using a first-order, multiphase decay model (Afvalzorg) containing data mainly for waste categories with low organic carbon content. The Afvalzorg model was chosen because previous studies found that this model best represented the conditions most prevalent at Danish landfills (Mou et al., 2015). The input data for the model were annual landfilled waste amounts distributed across eight waste categories. The modelled CH_4 generation was compared to an estimated CH_4 generation based on pumping tests from extraction wells installed in the waste body of the landfill. For more details about input data for the modelled CH_4 generation and the assumption for the estimated CH_4 generation from pumping tests, see paper **II**.

An average CH_4 recovery rate was calculated from the annual amount of recovered CH_4 reported by the operator, assuming evenly distributed gas recovery throughout the year.

Long-term changes in CH_4 storage were assumed not to influence the CH_4 mass balance in the relatively short time horizon used in this study (2013-2015). Changes in CH_4 storage resulting from changes, for example, in barometric pressure were assessed using the ideal gas law, to influence only the short-term changes of hours or days and not to affect the CH_4 mass balance over a period of three years (see paper **II**).

The remediation systems pump migrating landfill gas continuously from the soil formation around Hedeland landfill. Concentrations of CH_4, CO_2 and O_2 in the off-gas from the remediation systems and in monitoring wells have been measured regularly for many years. From the pumping rates and the measured CH_4 concentrations, the amount of CH_4 migrating from the landfill was calculated. A carbon mass balance based on the gas composition of the raw LFG and the off-gas from the remediation systems was used to estimate the initial amount of CH_4 laterally migrating from the landfill, before any oxidation took place in the soil compartment adjacent to the site. More details on the estimation of laterally migrating landfill gas are available in paper **II**.

Methane emissions were quantified by using the tracer gas dispersion method. Four measuring campaigns were conducted from 2013-2015 to investigate CH_4 emissions from Hedeland landfill under different conditions, in order to understand better emission patterns. Details about the measuring conditions are available in paper **II**.

Methane oxidation in the cover soil for a whole landfill site can be estimated using stable carbon isotopic analysis. The method is based on the fact that methanotrophs prefers ^{12}C over ^{13}C oxidising ^{12}C at a slightly higher rate than ^{13}C, resulting in the remaining CH_4 becoming ^{13}C-enriched. The method was applied once at Hedeland landfill, and samples for the analysis were collected at the site in May 2016. All gas samples were analysed at Florida State University. More details on the samples and the method are available in paper **II**.

2.3 Screening tool for locating landfill gas emission hotspots

To test a UAS-mounted TIR camera's ability to delineate LFG emission hotspots, a TIM 450 with a 38° aperture angle lens (Micro Epsilon, Germany) was mounted on an octocopter. To test the system, two 100 m^2 test areas were established at two landfills (Hedeland landfill and Audebo landfill), where 100 and 101 measuring points, respectively, were established (see paper **I** for details about the test areas).

At Hedeland landfill, four measuring campaigns were conducted during December 2015, and at Audebo landfill five measuring campaigns were conducted during March 2016. For all measuring campaigns, a thermal image of the soil surface in the test area was captured in the morning, before sunrise, and followed by measurements of CH_4 and CO_2 fluxes in the 100/101 measuring points together with measurements of soil temperatures on the surface and at 5- and 10-cm depths. More details on the methods employed for the flux and temperature measurements are available in paper **I**.

2.4 Landfill gas management in a biofilter

To test the potential of biofiltrating diluted landfill gas under field conditions, a pilot-scale compost-based biofilter was constructed at Hedeland landfill in a 30 m^3 open container. A 30-50 cm gas distribution layer (approximately 4 m^3) was constructed from pea gravel (8-16 mm) with a 110-130 cm compost layer (approximately 14 m^3) on top, making in total an 18 m^3 biofilter. The biofilter had a surface area of 11 m^2. Six horizontal multi-port gas samplers (HMPGSs) with nine ports each were installed over the depth of the filter, in order to monitor the gas composition inside the compost (see Figure 2). Combined moisture and temperature sensors connected to a data logger were installed at three depths at two locations in the filter. Detailed descriptions of the construction of the filter are available in paper **III**.

Raw LFG was collected from a nearby gas well installed in the waste body and mixed with air to a CH_4 concentration between 5 and 10 vol.%, to simulate diluted LFG.

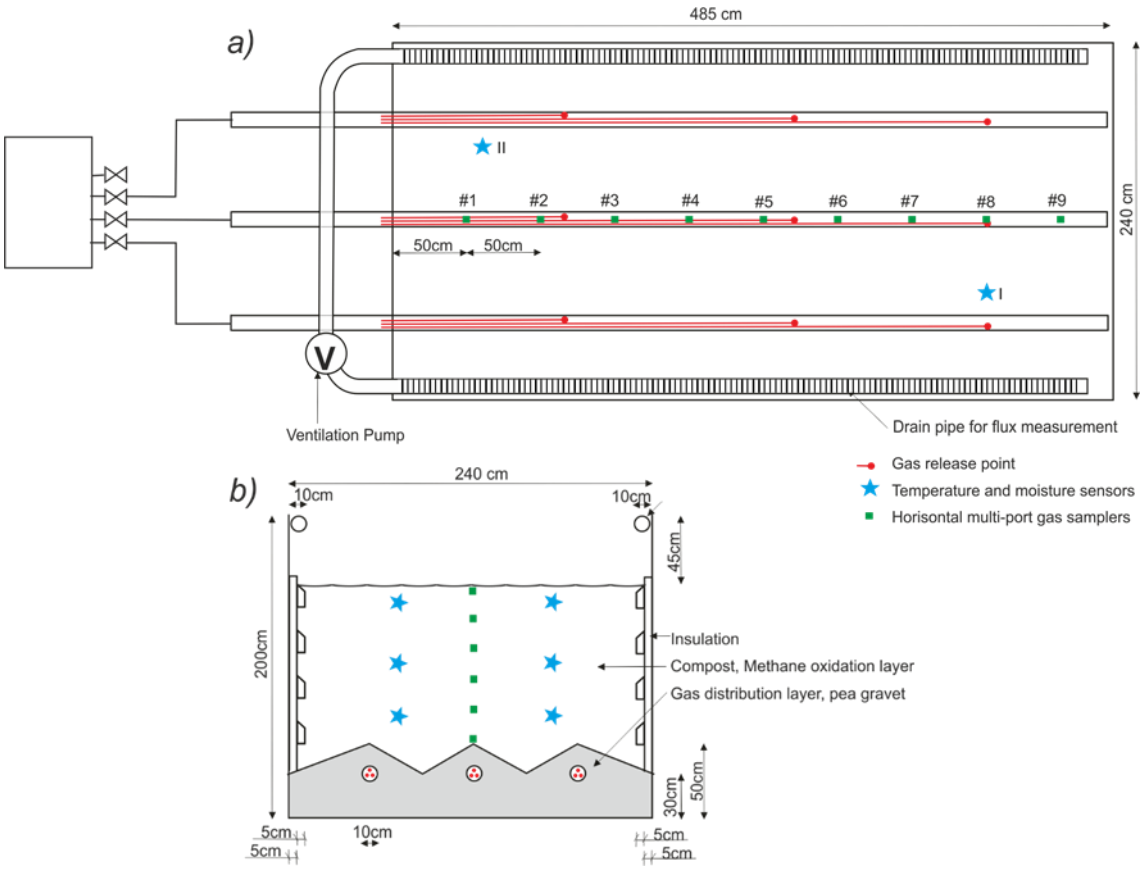

Figure 2. Principle outline of the pilot-scale biofilter. a) Filter seen from the top and b) a cross-section. From paper III.

The biofilter was tested in five flow campaigns, where the aim was to have the same inlet CH_4 concentration through all five flow campaigns but an increase in the load of CH_4. In addition, some background measurements were conducted before any CH_4 was loaded into the filter. Prior to initiating the five flow campaigns, background measurements were conducted in three flow campaigns – one with no load into the filter and two with just air loaded into the filter at two different flow rates.

Several measurement campaigns were conducted for each flow campaign, to document and follow the development of the filter performance, and on each measuring campaign gas composition was measured in all HMPGS ports together with gas concentrations of the inlet gas and the flux of CH_4 and CO_2 from the top of the filter. To measure the total flux of gases emanating from the top of the filter, a large flux chamber covering the whole container was constructed with a tarpaulin on top of the container and a flexible system for

securing this tarpaulin to the sides for flux measurements. CH_4 oxidation rates were calculated from a CH_4 mass balance of the filter.

A non-degradable HFC gas ($C_2H_2F_4$ or the more common HFC-134a) was used as a tracer gas, to test gas distribution in the filter and to support qualitatively the methane oxidation results obtained from the gas profiles in the HMPGS.

3 Landfill methane mass balance

Figure 3 presents the modelled CH_4 generation for Hedeland landfill, using the Afvalzorg model, and shows that CH_4 generation peaked in 1989-1991 and has declined since, albeit with a slight increase in 2006-2010, which was probably due to a significant rise in landfilled waste amounts in the last couple of years prior to the operation's termination in 2009. Using the Afvalzorg model, CH_4 generation was determined at between 57 and 79 kg h^{-1} in 2013-2015.

As part of ongoing investigations at the landfill, five large gas wells were installed in the waste body. LFG was extracted from each of the five wells successively. The purpose of these pumping tests was to see if it was possible to prevent LFG from migrating to the neighbouring plots if the gas was extracted from the waste body in the areas where migration had been identified. Data from these pumping tests included pump flows and gas concentrations of CH_4, CO_2 and O_2. In each test, LFG was extracted from the pumping well for about two months, after which the concentration of CH_4 in the well seemed to stabilise. Assuming that this stabilised concentration of CH_4 was an indication of the level of its generation in the area affected by the pumping well, a CH_4

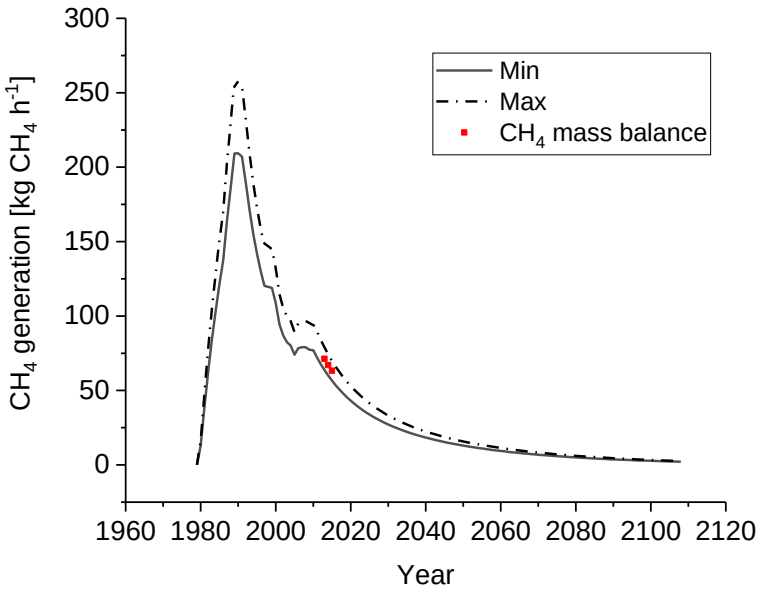

Figure 3. Methane generation for Hedeland landfill, modelled using the Afvalzorg model. Amended from paper II.

generation rate was calculated from the pumping rate and the gas concentration. The results from the pumping tests were used to estimate total CH_4 generated in the landfill waste. The influence radius of one out of the five pumping wells was assessed to overlap with two of the other pumping wells, so data from that well were excluded from the estimated CH_4 generation rate. The pumping tests were estimated to cover between 44 and 71% of the total landfill waste volume, and based on this premise and the measured CH_4 flow rates, the total CH_4 generation was estimated at 92-148 kg h^{-1} (assuming the same CH_4 generation rate for the whole landfill body). See paper **II** for further details on CH_4 generation.

The annual amount of CH_4 recovered from the landfill in 2013, 2014 and 2015 was reported by the engine operator. LFG recovery was assumed to be evenly distributed over the year, resulting in an average recovery rate of 12, 6 and 8 kg CH_4 h^{-1}, respectively. The large variation in the recovered amounts was due to the engine being out for service for several months in both 2014 and 2015.

The results of the CH_4 emission measurements showed varying emission rates between 3.1 ±0.7 and 13.4 ±0.8 kg h^{-1}, with the lowest emission rate of 3.1 kg h^{-1} occurring when the gas recovery system was running, while the highest emission rate of 13.4 kg h^{-1} was when gas recovery was stopped for 15 weeks prior to the measurements.

In the three remediation systems, 0.8 kg h^{-1} of CH_4 was captured in 2015. Analysis of the measured CH_4 and CO_2 concentrations in the monitoring wells showed indications of CH_4 oxidation in the soil compartment adjacent to the landfill. A carbon mass balance for the remediation of off-gas showed that 3.5 kg h^{-1} of CH_4 was initially migrating from the landfill, of which 2.7 kg h^{-1} was oxidised in the soil compartment. Extrapolating these results to the whole perimeter of the landfill resulted in a total rate of CH_4 laterally migrating in the range of 6.9 to 10.4 kg CH_4 h^{-1}, including the amount of CH_4 oxidised in the unsaturated zone before registration in the monitoring wells. Details about the assumptions behind the extrapolation of the results are available in paper **II**.

The results of the stable carbon isotope analyses for Hedeland landfill showed significant variations, resulting in the fractionation of oxidised CH_4 of 12, 27 and 92% for the three analysed downwind samples, respectively. With an average CH_4 emission rate of 8 kg h^{-1}, these three fractionation factors resulted in CH_4 oxidation rates of 1, 3 and 92 kg h^{-1}, respectively. CH_4 oxidation in the cover soil was thought to be the term of the CH_4 mass balance with the highest uncertainty in the determination of the rate. Several observations supported a

high oxidation rate in the cover soil. Surface screening with an FID showed very limited hotspots with elevated CH_4 concentrations, defined as concentrations above 25 ppmv CH_4 at the surface (see a detailed map with screening results in the supporting material to paper **II**). Detailed investigations of surface fluxes of CH_4 and CO_2 in the test area, described in section 2.3 and section 4, showed a fractional CH_4 oxidation of 79%, using a carbon mass balance based on the gas composition of the raw LFG and the measured surface fluxes.

The overall distribution of the generated CH_4 between the individual migration pathways is shown in Figure 4. Excluding CH_4 oxidation in the cover soil, and using average rates for the other terms of the CH_4 mass balance, resulted in a deficit in the mass balance of 42 kg h^{-1}, corresponding to 62% of the generated CH_4 and an oxidation rate in the cover of about 20 g m^{-2} d^{-1}, assuming only half of the cover is active in the oxidation (50,000 m^2). A review study found oxidation rates in the range of 22 to 230 g CH_4 m^{-2} d^{-1} in column tests simulating landfill soil covers (Scheutz et al., 2009), thus indicating that it is reasonable to assume that CH_4 oxidation in the cover can close the CH_4 mass balance for Hedeland landfill. Even with a higher CH_4 generation resulting in a higher deficit in the mass balance (see Table 1, where the results of the mass balance is shown using the CH_4 generation estimated from the pumping test together

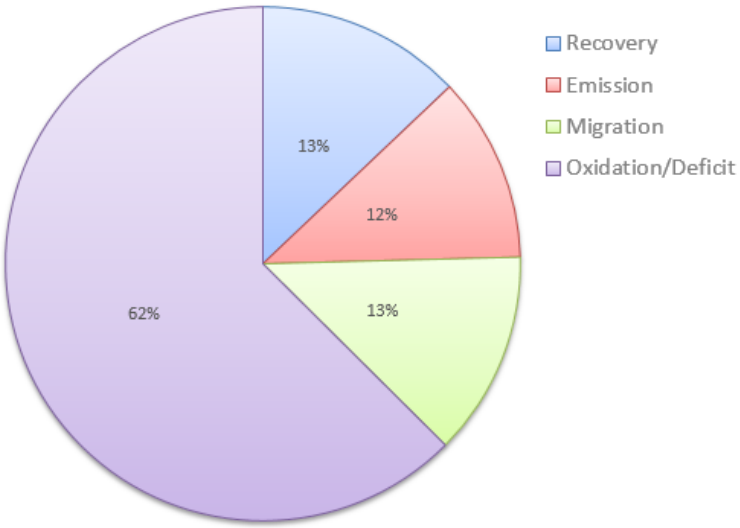

Figure 4. Results of CH_4 mass balance for Hedeland landfill, showing the distribution of the generated CH_4 between the individual migration pathways. Migration represents only laterally migrating CH_4, including what was oxidised in the soil compartment adjacent to the landfill.

with the modelled CH_4 generation). A deficit of 95 kg h^{-1} corresponds to a load to the cover of about 45 g m^{-2} d^{-1}, with the same assumption of half of the cover being active for oxidation, and it is still at the lower end of the results found in the previously mentioned review study.

Annual average CH_4 oxidation was established at 89% in a sandy soil for laterally migrating LFG (Christophersen et al., 2001), which was the highest fraction reported by Chanton et al. (2009) for field studies in a review of 42 determinations of the fraction of oxidised CH_4. The review details studies from many different landfills with a variety of covers and soil types, from a range of different countries, and looks at both in situ field studies and laboratory column tests of landfill cover materials. The fractions of oxidised CH_4 found in these field studies varied between 0.01 and 0.89 (Chanton et al., 2009), with the average from five field studies conducted in Northern Europe, which should be comparable to the ambient conditions at Hedeland landfill, at 54 ±14% (Chanton et al., 2009). Only four out of the 42 determinations reported a value of 10% CH_4 oxidation or less (Chanton et al., 2009), which indicates that the default value of 10% suggested by IPCC for national inventory reporting (IPCC, 2006) could be significantly underestimating CH_4 oxidation at many landfill sites.

Several circumstances can influence oxidation efficiency in the landfill cover, for instance contact between LFG and methanotrophs in the soil, meaning that if LFG is emitted into the atmosphere, such as from installations in the landfill in the form of leachate wells, the overall oxidation efficiency of the site can be low, even with high local oxidation efficiencies in the cover. The leachate

Table 1. Methane mass balance for Hedeland landfill for modelled CH_4 generation and CH_4 generation determined from pumping tests. All results are averages, and units are in kg h^{-1}. Numbers in brackets give the distribution in percentages. Amended from the Supporting Material to Paper II.

CH_4 generation	**CH_4 recovery**	**CH_4 emission**	**CH_4 lateral migration[1]**	**CH_4 oxidation in cover**	**Deficit excl. oxidation in cover**
Modelled					
67	9 (13%)	8 (12%)	9 (13%)	32 (48%)	*42 (62%)*
Pumping tests					
120	9 (7%)	8 (7%)	9 (7%)	32 (27%)	*95 (79%)*

[1]Including oxidation of laterally migrating LFG

collection system was found to contribute approximately 47% and 27% of the total LFG emissions, respectively, at two Danish landfills (Fredenslund et al., 2010; Scheutz et al., 2011b). At Hedeland, only one leachate well drains all units of the landfill, and measurements have shown that emissions from that well are limited, thus supporting the assumption that the main part of the generated LFG is loaded to the cover.

Hedeland landfill was established in an abandoned gravel pit, and so the adjacent soil compartment is constituted by relatively dry sand and gravel, where little methane oxidising microbial activity is expected. Nevertheless, indications of methane oxidation were found along the whole perimeter of the landfill, with elevated CO_2 concentrations in nearly every monitoring well. Though the oxidation rate is small, it can still be significant due to the size of the soil compartment with a gravel layer of 10-15 metres.

A CH_4 recovery efficiency of only 13% indicates a high potential for optimising LFG collection, but an extension to the existing gas collection and recovery system will require installing new and expensive gas extraction wells, while at the same time CH_4 content in the landfill is declining. Furthermore, the emission rate at Hedeland landfill was at the low end compared to similar landfills. The measured emission rate was in the lower half compared to 14 other Danish landfills, where emission rates between 2.6 and 60.8 kg CH_4 h^{-1} were found (Mønster et al., 2015) and in most cases were significantly lower than the 12.8 to 441 kg CH_4 h^{-1} reported for seven Swedish landfills (Börjesson et al., 2009).

Lateral migration of LFG is often not quantified either, because this migration pathway was found not to be relevant due to the design of the landfill, as it is only relevant for landfills constructed partly underground or it could be because bottom and side liners were installed to prevent gas and leachate migration into the surroundings. Therefore, lateral migration is most often either calculated as diffusional transport through an uncompromised liner or neglected entirely, because it is assumed to be insignificant in comparison to the other migration pathways. The size of the laterally migrating CH_4 was found to be as large as the recovered CH_4 and CH_4 emissions from Hedeland landfill. The results show that the liners were not uncompromised, thus the lateral migration was not insignificant and could not be neglected. Bottom and side liners at Hedeland landfill were clearly constructed to protect the environment against seeping leachate, and the generation and migration of LFG was not a consideration. The side liners were constructed with no overlapping connections, as

can be seen in Figure 1, thereby creating weak areas where cracks in the clay liner would present an easy escape pathway for the LFG. This could also be the case at other landfill sites, as nothing indicates that something exceptional should have occurred at Hedeland.

4 Screening tool for locating landfill gas emission hotspots

A UAS-mounted TIR camera's ability to delineate LFG emission hotspots relies on a significant correlation between surface soil temperatures and emission rates. To determine if a correlation could be found, surface temperatures obtained with a TIR camera and with a thermometer inserted into the soil were compared to surface fluxes of CH_4 and CO_2 at two Danish landfills in test areas established where FID surface screenings had shown elevated CH_4 concentrations.

Changes in atmospheric pressure affect LFG migration and emissions (e.g. Aghdam et al., 2018; Czepiel et al., 2003; Xu et al., 2014), so the average CH_4 fluxes from each of the measuring campaigns (obtained from interpolating all flux measurements using MapInfo Vertical Mapper (natural neighbour)) were plotted against changes in the atmospheric pressure (see Figure 5). The results indicated that at Hedeland landfill CH_4 fluxes were highly influenced by changes in atmospheric pressure, as none was measured while atmospheric

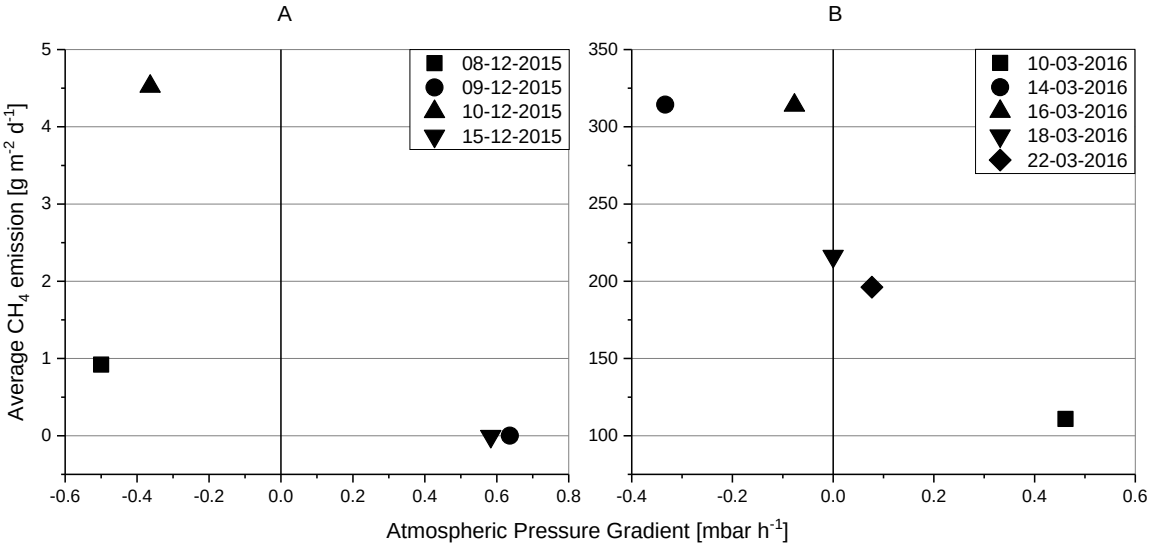

Figure 5. Average CH_4 fluxes versus atmospheric pressure gradient at A: Hedeland landfill and B: Audebo landfill. From Paper I. The average flux was determined by interpolating the flux measurements using MapInfo Vertical Mapper and then calculated the average from the interpolated data. Please note the different scales on the y-axis.

pressure was increasing, and only minor fluxes were measured at the days with a decrease in atmospheric pressure. At Audebo landfill, the same tendency was observed, but in general the average flux was significantly higher than at Hedeland. Furthermore, at Audebo, there was still an observable CH_4 flux, even with an increase in atmospheric pressure, albeit with a lower rate compared to when atmospheric pressure was decreasing.

Methane fluxes at Hedeland landfill varied between -1.9 and 290 g m^{-2} d^{-1}, with an average of 1.3 ±16 g CH_4 m^{-2} d^{-1} for all four measuring campaigns, while CO_2 fluxes varied between <0.05 and 184 g m^{-2} d^{-1} and an average of 16.4 ±28 g m^{-2} d^{-1}. In the month prior to the measurements at Hedeland, the level of precipitation was much higher than the usual average for that region and month of the year. The higher level of precipitation was thought to have saturated the soil pores with water, thus blocking gas migration and being the reason for the relatively low emissions measured, compared to the indications from previous investigations in the same area, showing emission hotspots with elevated CH_4 concentrations. Comparing the soil temperatures measured at a 10-cm depth, the surface temperatures obtained from the TIR images and the measured flux of CH_4 and CO_2, showed limited correlations for Hedeland landfill (see Figure 6, showing data from 10 December 2015, the measuring campaign where the highest CH_4 fluxes was registered). It is important to note that the test area at Hedeland was located on a slope with a plateau at the top, just above the test area. The TIR images were captured from an opposite slope, resulting in the test area and the plateau having two different angles to the TIR camera, following which temperatures for the two areas were not comparable (the warmest area in the western part (top) of the TIR images from Hedeland landfill being the plateau). Figure 7 illustrates the setup with the TIR camera on the slope opposite to the test area.

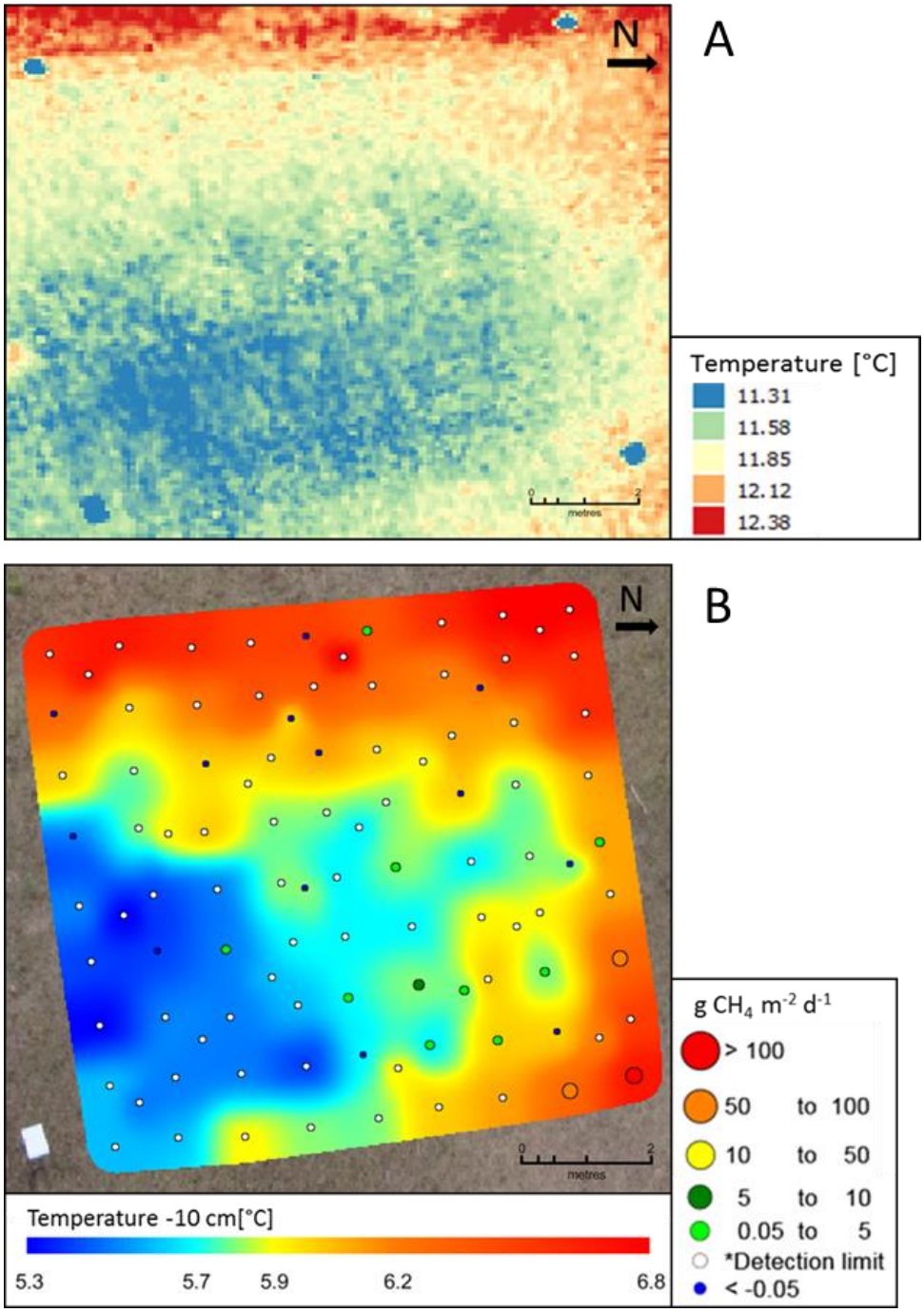

Figure 6. Results from Hedeland landfill, 10 December 2015. A: TIR image of the test area. Cold areas (blue dots) in the corners are ground control points (40x40 cm aluminium plates) marking the corners of the test area. The test area was located on a slope, and the TIR image was captured from an opposite slope. The warmer area at the top of the image is a plateau above the slope, at another angle to the camera, and thus the temperature in that area is not comparable to the temperature in the test area. B: Interpolated temperature measured at a depth of 10 cm, with coloured dots representing CH_4 surface fluxes. *Detection limit of flux measurements was 0.05 g m^{-2} d^{-1}. Amended from paper I.

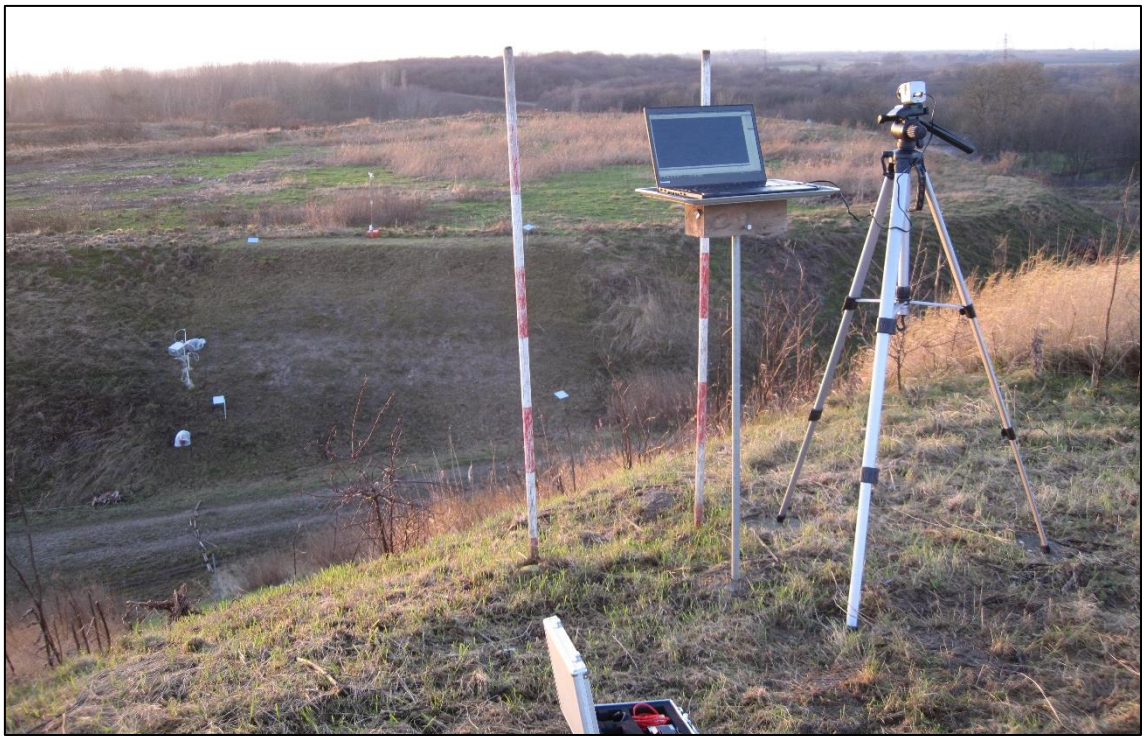

Figure 7. Picture illustrating the setup with the TIR camera at Hedeland landfill and showing the plateau above the test area at another angle to the TIR camera, making other surface temperatures incomparable to the surface temperatures obtained for the test area.

Methane measurements at Audebo landfill showed CH_4 fluxes between -22.7 and 14,572 g CH_4 m^{-2} d^{-1}, with an average of 371 ±1337 g CH_4 m^{-2} d^{-1} based on five measuring campaigns. CO_2 fluxes were between <0.05 and 45,240 g CO_2 m^{-2} d^{-1}, with an average for all five measuring campaigns of 860 ±3614 g CO_2 m^{-2} d^{-1}. Soil temperatures measured at 10 cm in depth, surface temperatures obtained from the TIR images and measured fluxes of both CH_4 and CO_2 showed the same patterns in all datasets with higher temperatures, both at 10 cm and on the surface in the same area, as the highest LFG fluxes were measured (see Figure 8, showing data from Audebo landfill 14 March 2016). Results from the rest of the measuring campaigns from both Hedeland and Audebo can be found in the supplemental material for paper **I**.

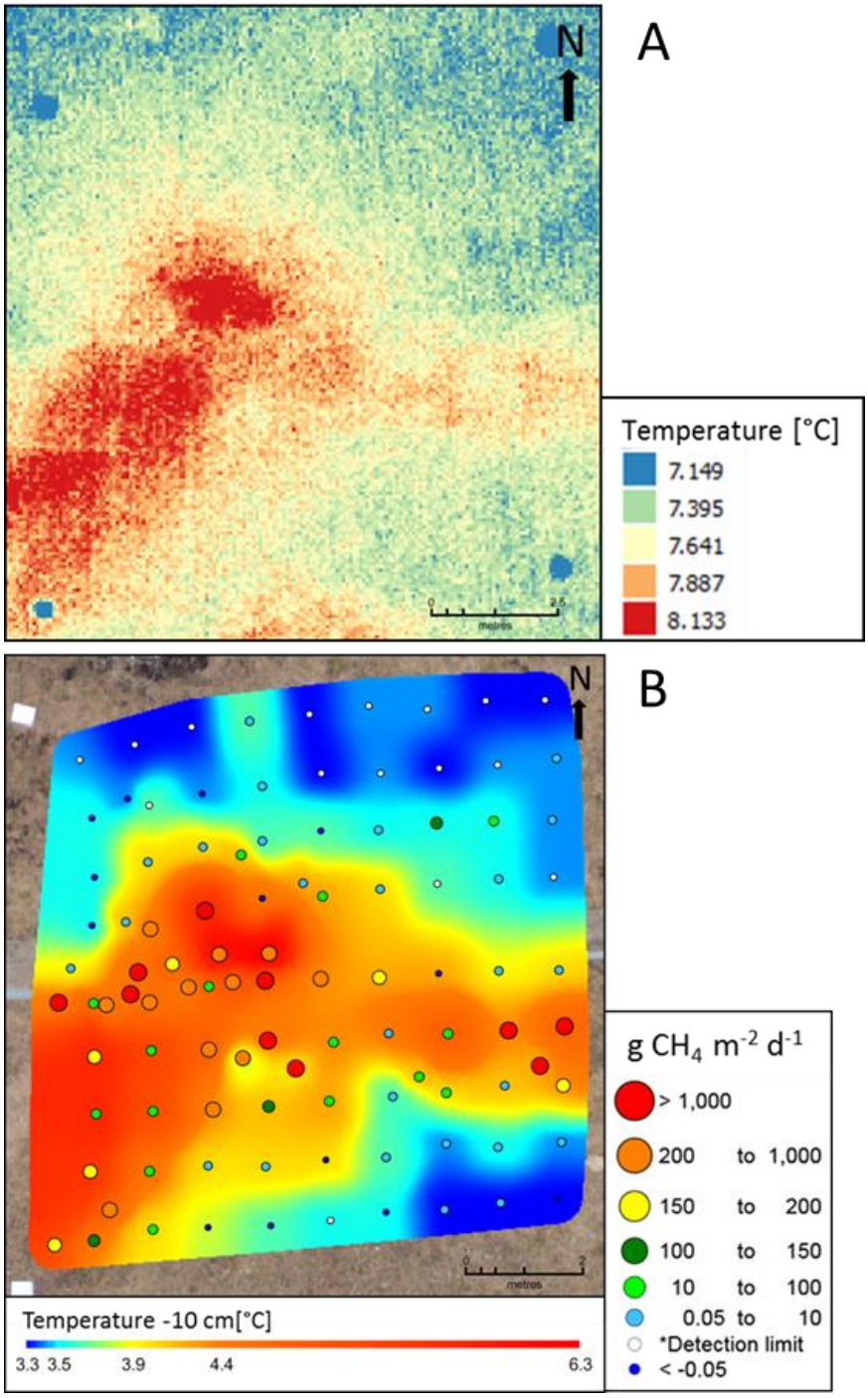

Figure 8. Results from Audebo landfill, 14 March 2016. A: TIR image of the test area. Cold areas (blue dots) in the corners are ground control points (40x40 cm aluminium plates) marking the corners of the test area. B: Interpolated temperature measured at 10 cm depth, with coloured dots representing CH_4 surface fluxes. *Detection limit of flux measurements was 0.05 g m^{-2} d^{-1}. Amended from paper I.

Maximum temperature differences on the surface inside the test areas obtained from the TIR images were about 1°C at both Hedeland and Audebo, whereas the temperature difference at 10 cm in depth was up to about 2°C at Hedeland landfill and up to 5°C at Audebo landfill. This indicates that the temperature differences were less pronounced on the surface than at a 10 cm depth, especially for Audebo. One reason for this result could be that temperatures reported by the TIR camera are influenced by the emissivity of the materials or objects in the image. Vegetation, for instance, will have emissivity different to a bare soil surface, so because of vegetation the TIR camera was probably not always reporting the temperature of the soil but of the vegetation, which most likely was influenced more by the temperature of the ambient air than the temperature of the emitting LFG.

Other studies using a TIR camera for delineating LFG emission hotspots have reported temperature differences as high as 30°C, but more commonly they are in the range of 2 to 15°C, with a few cases of higher differences (see Table 1 in paper **I**). An explanation for the higher temperature differences reported in the literature could be due to the type of landfills studied. Landfill waste temperatures of up to 65°C have been reported (Hanson et al., 2010), and the temperature is expected to be higher at sites with a high content of easily degradable organic matter, such as municipal solid waste (MSW), than at locations with mainly non-combustible waste. There has been a ban in Denmark since 1997 on landfilling waste suitable for incineration, and so waste landfilled at Danish sites has a low organic carbon content, which could result in lower gas temperatures.

Some of the highest CH_4 fluxes measured at Audebo landfill were measured at the emission hotspots named HS5, HS10 and HS14. Pictures of the hotspots, together with a zoom of the TIR images indicating the location of the respective hotspots in the image, can be seen in Figure 9 and Figure 10. These pictures show that even though the highest CH_4 fluxes were measured at these three hotspots, and they were significantly higher than the fluxes measured at the closest measuring points, they were not significantly distinguishable in the TIR images. For HS5, it was thought to be because the physical area covered by the hotspot would be represented in the TIR images by only a few pixels. For HS10 and HS14, it was thought to be because the exact emission of CH_4 happened from a hole either at the bottom of the bigger hole (HS10) or from the vertical part of the hole (HS14), so the actual emission hotspot was either not visible from the air (HS14) or the warm LFG had limited contact with the visible soil surface (HS10). This indicates that the method has some limita-

tions when it comes to delineating single hotspots with a limited area distribution.

Thus, the results indicate that a UAS-mounted TIR camera is capable of delineating LFG emission hotspots with a minimum flux of 150 g CH_4 m^{-2} d^{-1} from an area of at least 1 m^2.

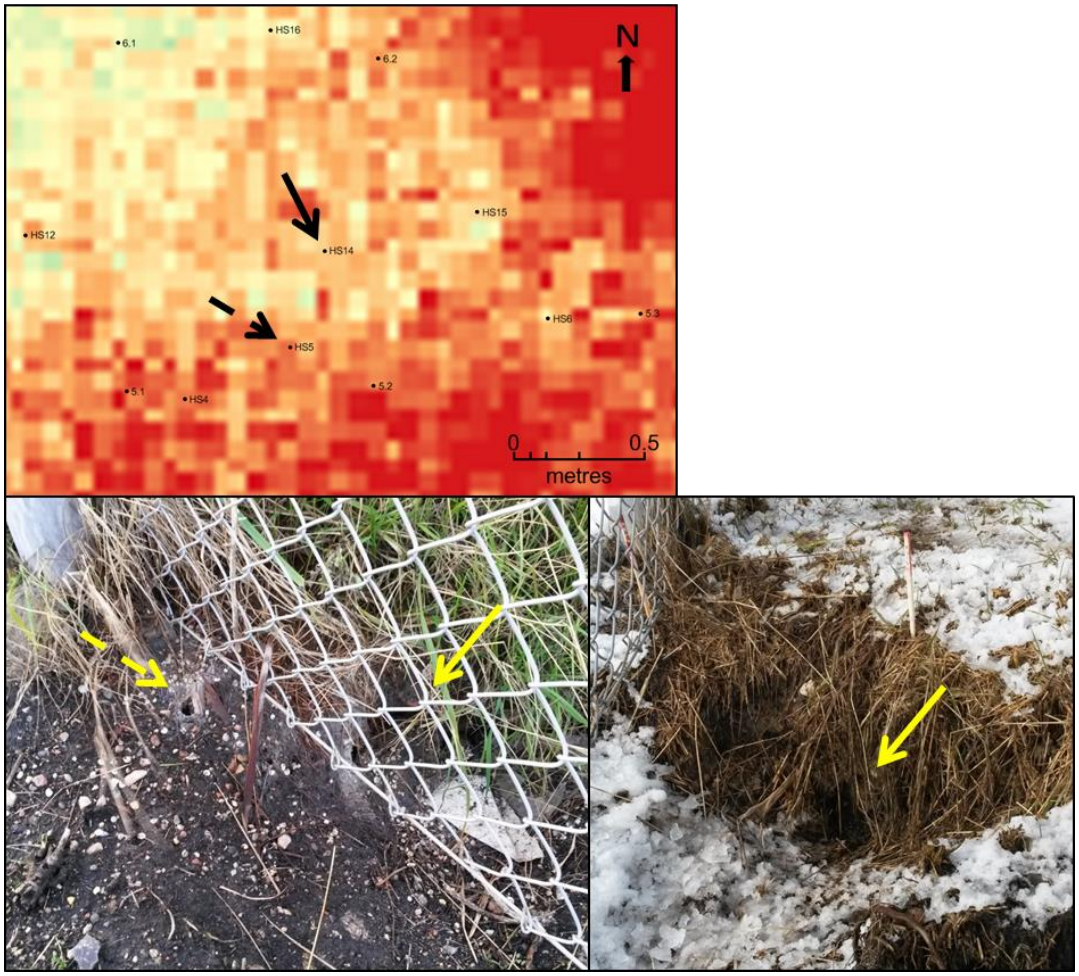

Figure 9. Emission hotspot HS14 (full line arrow) and HS5 (dashed line arrow) at Audebo landfill. TIR image from 14 March 2016. From paper I.

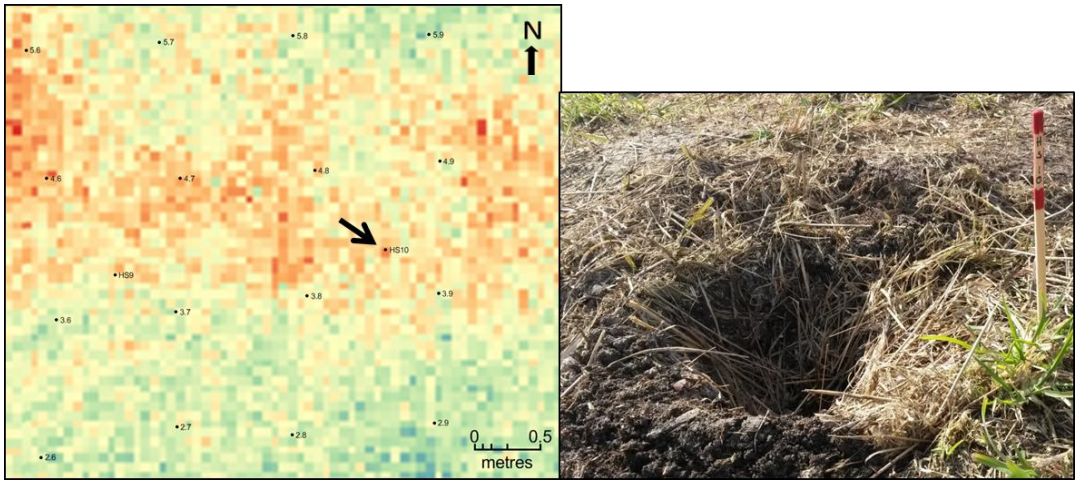

Figure 10. Emission hotspot HS10 at Audebo landfill. TIR image from 14 March 2016. From paper I.

5 Landfill gas management in a biofilter

The actively loaded biofilter for treating diluted landfill gas was tested in five flow campaigns with the same intended CH_4 inlet concentration and a flow rate resulting in an increasing CH_4 load into the filter. A CH_4 inlet concentration of 8 vol.% was targeted. Fluctuations in the CH_4 concentrations of the raw LFG, and in the pump flow rate, which was not completely stable, resulted in variations in inlet concentrations – and thus the CH_4 load into the filter. Furthermore, a generally decreasing concentration of CH_4 in the raw LFG was observed during the experiment (see Figure 11 and paper **III**).

The dispersal of gas in the distribution layer was tested by injecting a tracer gas (HFC-134a), with the results showing a fast and even spreading.

Background measurements were conducted before any LFG load was started. The initial measurements showed O_2 at an average concentration of 16.7 ±1.1 vol.% throughout the whole depth of the filter, and an average concentration of CO_2 of 5.1 ±1.1 vol.%. The reduced O_2 level and increased CO_2 level resulted from compost respiration, in both cases when there was no load into the filter. When air was loaded into the filter, O_2 concentration increased to an average level close to ambient conditions, while CO_2 concentrations decreased to an average of 2.4 ±1.4 vol.%. The background measurements were conducted in January, and even during the cold season, significant compost respiration was still measurable, judging from the CO_2 concentrations in the biofilter. In

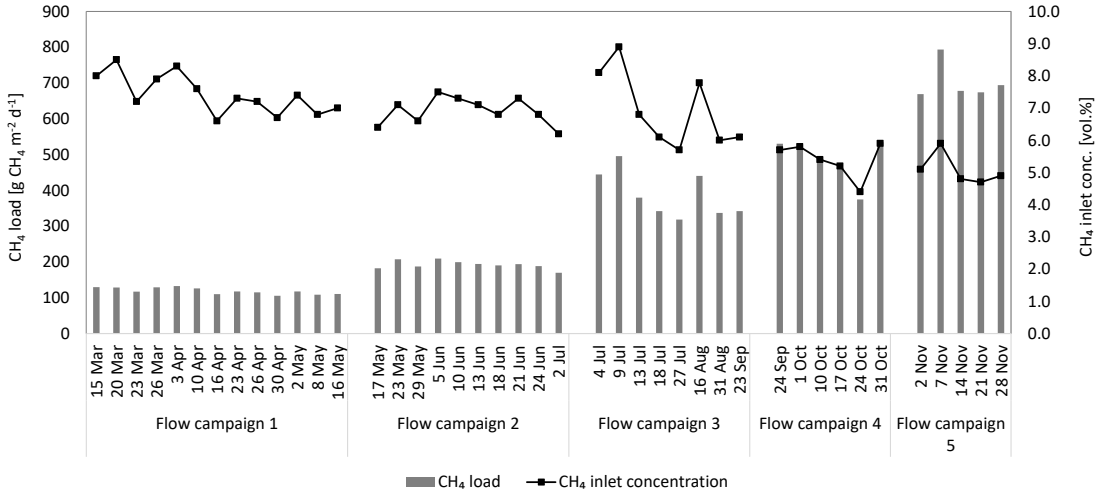

Figure 11. CH_4 inlet concentrations and CH_4 load into the biofilter through all five flow campaigns. From paper III.

addition, slightly elevated temperatures in the filter (up to 10°C above the ambient air) indicated microbial activity. Though, a batch test of the compost respiration showed O_2 consumption below the recommended upper limit.

At each measuring campaign, the gas distribution in all HMPGS ports was measured, and pore gas concentrations were plotted over the depth of the filter. Figure 12 shows a series of examples of gas profiles, two from each flow campaign, one approximately after 2 weeks at the given flow campaign and one at the end of the given flow campaign; all profiles are for HMPGS port #5. At the start of flow campaign 1, there was an initial lag phase of about 40 days before CH_4 oxidation started. The gas profiles for flow campaigns 2 and 3 show that during flow campaign 2 the filter was O_2-depleted and started producing CH_4 that continued at the beginning of flow campaign 3. The produced CH_4, however was oxidised in the upper part of the filter and did not seem to influence the overall efficiency of the filter. The anaerobic conditions in the middle of the filter were thought to be due to the thick compost layer (~1.2 m) and could most likely be avoided with a thinner layer of compost. Another solution could be multiple air injections in different levels of the filter, as demonstrated by Farrokhzadeh et al. (2017) and Haubrichs and Widmann (2006), activating even more of the filter material for CH_4 oxidation. Then again, as most of the methane was oxidised in the lowest 20 cm of the filter, additional air injections would only help prevent anaerobic conditions. Thus, multiple gas injections (with dilute landfill gas) could be a way of activating more of the filter volume, in order to both prevent anaerobic conditions and increase the oxidation rate of the filter.

Figure 13 shows CH_4 oxidation rates and oxidation efficiencies for all five flow campaigns. The rate was relatively stable during flow campaigns 1 and 2, with an initial lag phase lasting the first month of flow campaign 1, most likely due to adaptation of the methanotrophic culture. The average CH_4 oxidation rate was 68 ±19 g m^{-2} d^{-1} for flow campaign 1 and 140 ±22 g m^{-2} d^{-1} for flow campaign 2. Excluding the initial lag phase, the average CH_4 oxidation rate for flow campaign 1 increased to 80 ±7 g m^{-2} d^{-1}. Overall CH_4 oxidation efficiency increased during the first two flow campaigns from an average of 46 ±15% in the first month to above 80% in the last month of flow campaign 2, albeit with a decreasing tendency at the end of flow campaign 2. In flow campaign 5, an average CH_4 oxidation rate of 387 ±52 g m^{-2} d^{-1} was obtained with an overall CH_4 oxidation efficiency of 55 ±5%.

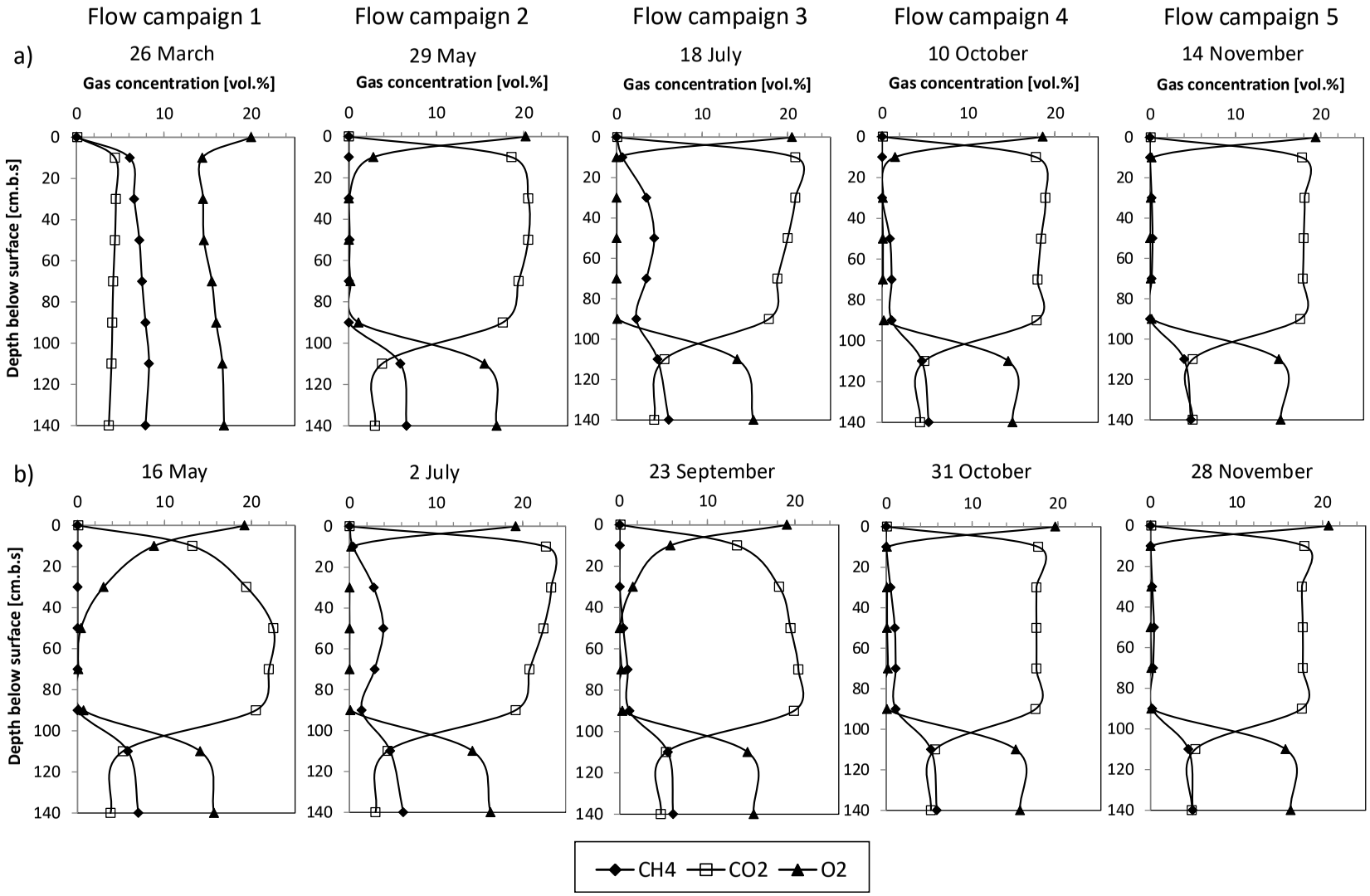

Figure 12. Gas concentration profiles for CH_4, CO_2 and O_2 measured in port #5 in all HMPGS a) after approximately two weeks of the given flow campaign and b) at the end of the given flow campaign. – Value at 140 cm.b.s. represents inlet concentrations and 0 cm.b.s. is ambient concentration. From paper III.

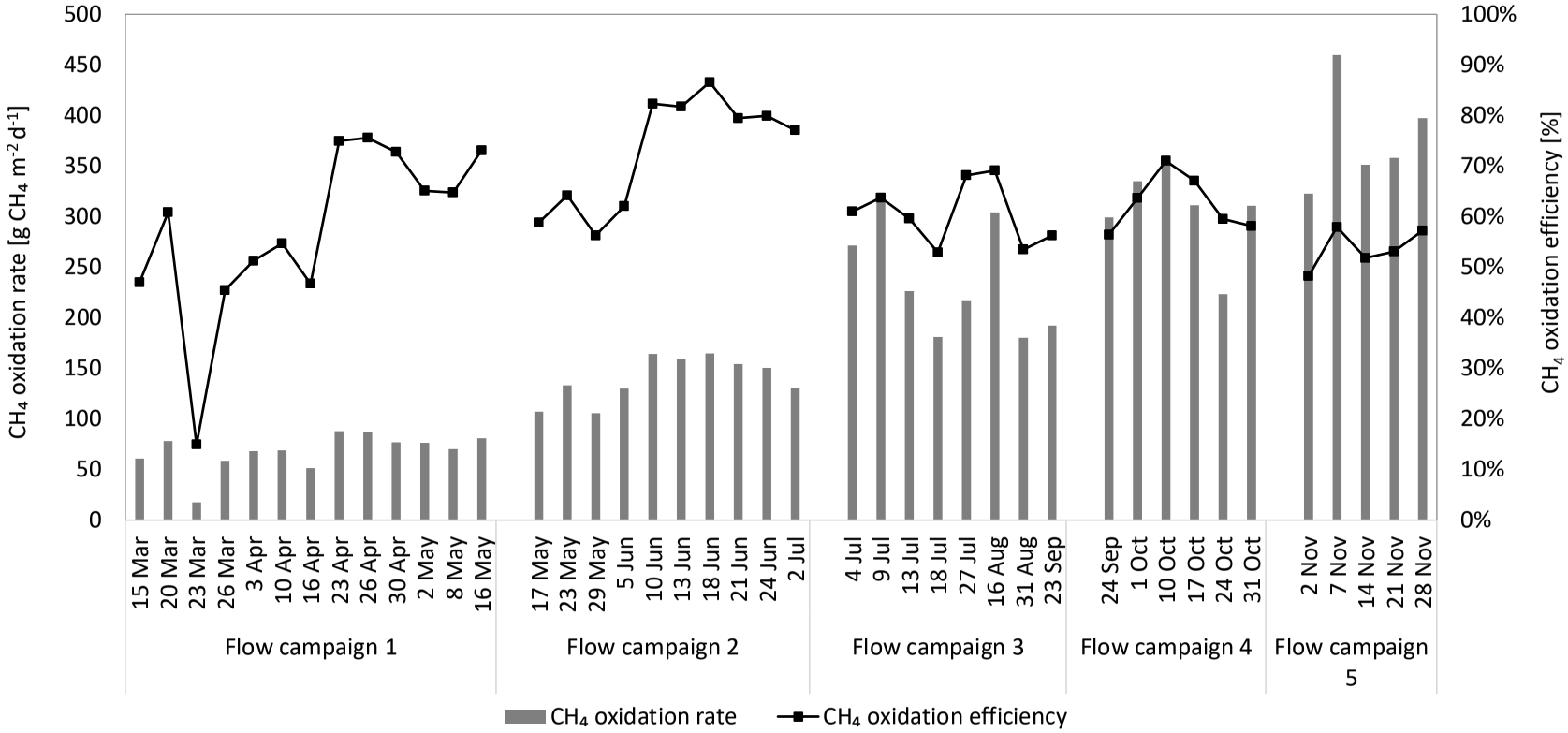

Figure 13. Methane oxidation rate and CH_4 oxidation efficiency. From paper III.

The low overall CH_4 oxidation efficiency was probably due to preferential flows. Surface screenings with a portable FID showed substantial preferential flows at the transition between the compost and the container wall, even though insulation plates of polystyrene with a jagged surface were glued to the container side, to insulate the container in the cold season and to prevent preferential flows. This construction turned out to be a rather short-lived solution, and despite several attempts to minimise the preferential flow, it did not last to the end.

The gas profiles obtained from the HMPGS showed CH_4 oxidation of almost 100% through all five flow campaigns for all HMPGS, albeit with a significant decrease in especially ports #1 and #9 in flow campaign 5. In Figure 14, showing the gas profiles for all HMPGS on the last day of flow campaign 5, it is clear that gas distribution and oxidation efficiency are different at ports #1 and #9 (located at each end of the biofilter). Tracer gas tests with HFC-134a supported the high oxidation rate in the HMPGS by comparing the ratio of CH_4 to HFC-134a in the individual ports of the HMPGS at 10 cm in depth to the ratio in the filter inlet. An average oxidation efficiency for all HMPGS was found at 86% for flow campaign 5 with a load of 694 g CH_4 m^{-2} d^{-1}. The ratio of CH_4 to HFC-134a in the lowest HMPGS, just above the gas distribution layer, showed that about 10% of the loaded CH_4 was oxidised in the gas distribution layer.

In a column test, packed with a similar compost as the one used in the biofilter at Hedeland landfill, a CH_4 oxidation rate of 509 g m^{-2} d^{-1} with an oxidation efficiency of almost 100% was obtained when the column was loaded with a CH_4 inlet concentration of 10 vol.% and mixed with air (Thomasen et al., 2019). The oxidation rate was similar to the highest of 460 g CH_4 m^{-2} d^{-1} found at Hedeland landfill with an CH_4 inlet concentration of about 5.9 vol.%, although efficiency was only 58%.

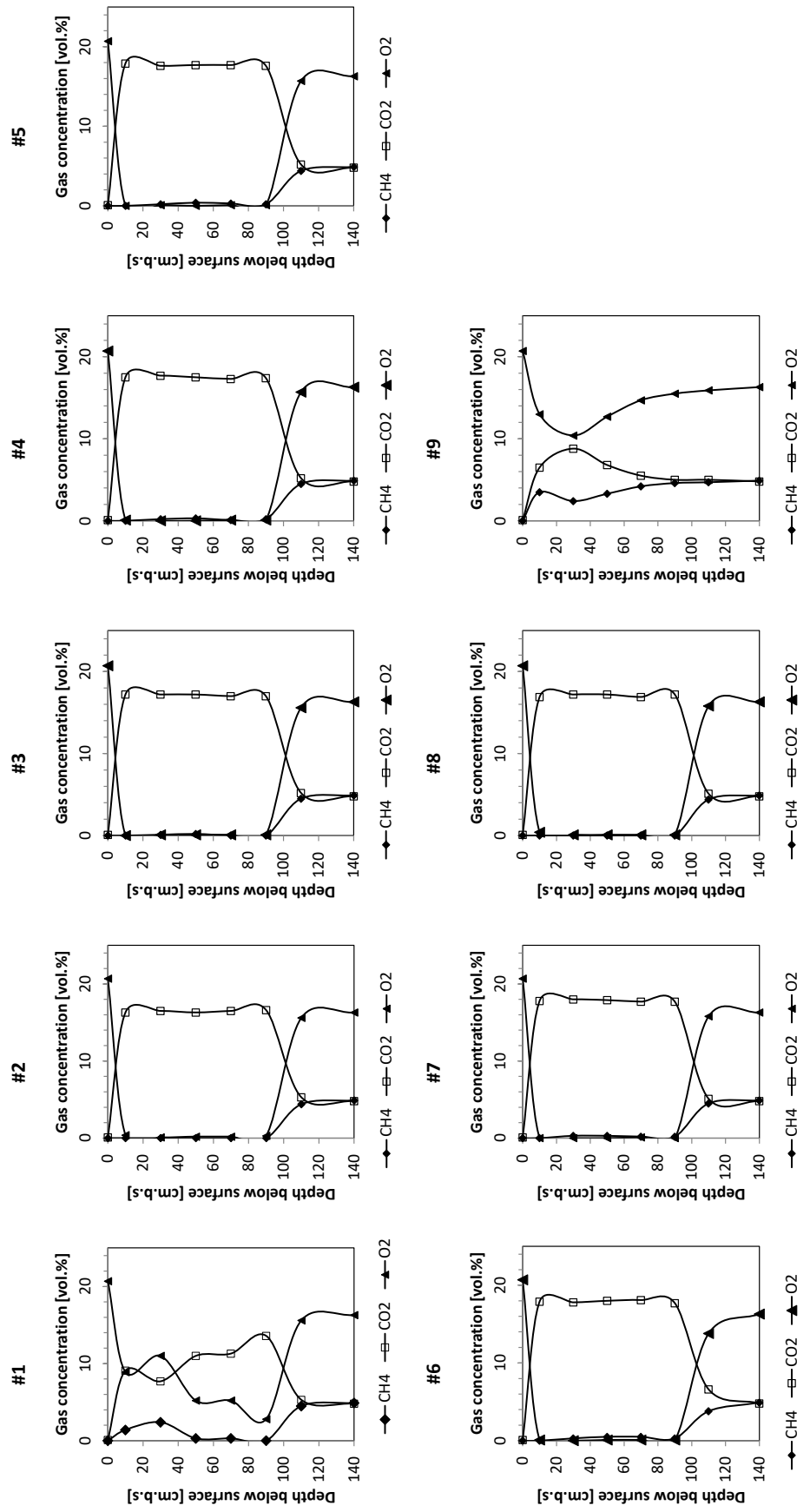

Figure 14. Gas profiles from all HMPGS from 28 November 2018 (the last day of flow campaign 5).

Figure 15 shows temperatures in the biofilter for all flow campaigns, including background campaigns, together with the ambient temperature, given as a daily average. A stable temperature between 0.9 and 2.6°C was observed in the compost the first two weeks of flow campaign 1 with ambient temperatures around freezing. Temperatures in the compost followed ambient temperatures for the first month of flow campaign 1. Temperatures in the filter continued to increase until a maximum temperature of between 52 and 59°C was obtained approximately four weeks into flow campaign 3. Temperatures in the filter stayed above 40°C to the end of flow campaign 5 and did not follow the decrease in autumn ambient temperatures. For more details on the temperature observations, see paper **III**. Streese and Stegmann (2003) observed similar temperatures (50°C) in their pilot-scale biofilter installed at a landfill, which resulted in the warm gas drying the filter material to an extent where adding extra water was necessary to maintain optimal conditions for microbial activity. However, their filter was constructed in the form of 12 biofilter units in four columns arranged in parallel, and precipitation probably did not infiltrate the biofilters. A relatively stable volumetric water content (VWC) between 0.4 and 0.6 m^3 m^{-3} was observed in the biofilter at Hedeland landfill despite high temperatures and a dry and warm summer with only minor rainfall recorded for two months. The VWC decreased slightly throughout flow campaigns 4 and 5 (in the autumn), even though rainfall occurred more often than during the summer period.

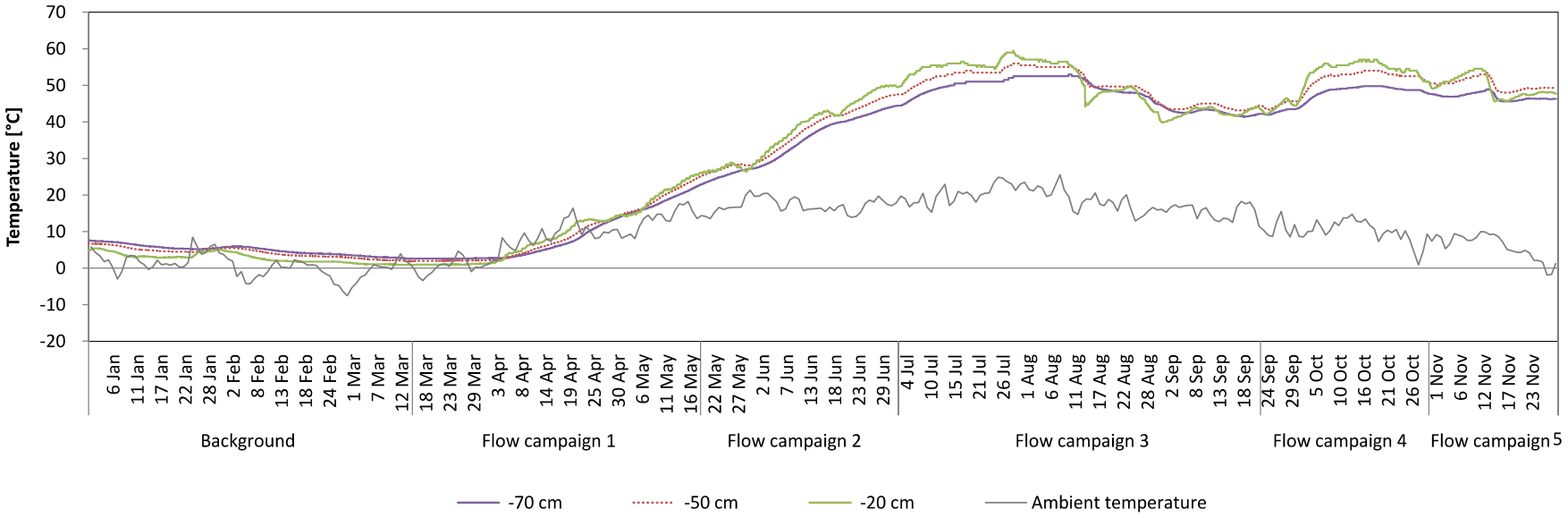

Figure 15. Compost temperatures logged at sensor group I (see Figure 2 for location) at three depths, together with ambient temperatures obtained as the daily average from a weather station approximately 1 km away from the landfill (DMI, 2018). Amended from paper III.

The competent authorities at Hedeland landfill require treatment of the off-gas from the three installed remediation systems, in order to protect the climate. An average CH_4 content of 0.53 ±0.55 vol.% was observed in 2017 from the three remediation systems, and O_2 content in most cases was above 10 vol.%. This would correspond to a CH_4 flow of 7,890 g CH_4 d^{-1} or a load of 717 g CH_4 m^{-2} d^{-1} if all the off-gas from the remediation systems were loaded into the constructed pilot-scale biofilter. With the actual flow of the remediation pumps (80 m^3 h^{-1} in total) it would result in a retention time of only 3 min, which is much lower than the critical retention time of 30-35 min suggested by Farrokhzadeh et al. (2017). About ten filters, the same size as the pilot-scale biofilter tested in this study, are required in order to treat off-gas from the three remediation systems with a retention time of at least 30 min. This will result in a load per filter of only 71 g CH_4 m^{-2} d^{-1}, which is much lower than the maximum oxidation rates found in this study – and only slightly higher than the design criteria used for passive biocover systems in the Danish Biocover initiative (Danish EPA, 2019). An alternative to multiple filters in containers could be a biofilter embedded into the landfill cover with a surface area of about 111 m^2, which would result in the same load per m^2 as ten containers. This solution is similar to the pilot-scale biofilter established at AV Miljø Landfill (Cassini et al., 2017; Scheutz et al., 2017) and could most likely also help solve the problem with preferential flows at the edges.

Another alternative would be to redesign the biofilter so that the diluted LFG is injected at different levels into the filter in addition to the bottom (not just air injected at multiple levels, as reported in other studies), which would increase the retention time and therefore require less filter volume. At the same time, more of the filter material could be expected to be utilised for CH_4 oxidation, in which case possibly higher CH_4 oxidation rates and efficiencies could be obtained.

6 Conclusions

The objective of the PhD project presented in this thesis was to establish and close the methane mass balance for Hedeland landfill, containing mainly non-combustible waste, to test a UAV-mounted thermal camera's ability to delineate landfill gas emission hotspots and to develop a compost-based mitigation technology for handling diluted landfill gas. The objectives were achieved through the detailed analysis of data from many years of investigations and by field investigations and demonstration tests at a landfill site. The main findings of the conducted research can be summarised as follows:

- Methane oxidation in the landfill cover could close the CH_4 mass balance for Hedeland landfill. About 38% of the total modelled CH_4 generation was accounted for by recovery, lateral migration and emission into the atmosphere, and the remaining 62% was covered by CH_4 oxidation in the cover, which is much higher than the 10% that is usually recommended as a default value for landfill cover oxidation. Landfill gas migration pathways are site-specific, and thus detailed site investigations are required to determine the individual migration pathways governing the overall CH_4 mass balance, which can then form the basis for a gas mitigation strategy.
- A UAS-mounted TIR camera could work as a cost-efficient screening tool to delineate landfill gas emission hotspots at Danish landfills under certain conditions. An emission of at least 150 g CH_4 m^{-2} d^{-1}, and with a spatial distribution of at least 1 m^2, is required for the TIR camera to identify emission hotspots. The method seems to be more successful at sites with a higher CH_4 generation rate and emission rate.
- Microbial CH_4 oxidation of diluted landfill gas was possible in a pilot-scale compost-based biofilter, where the highest oxidation rate found was 460 g CH_4 m^{-2} d^{-1} with an efficiency of 58%. CH_4 oxidation efficiencies above 90% were found from gas concentration profiles inside the compost. Overall efficiency was significantly reduced due to substantial preferential flows originating from a flawed construction method. Higher efficiency is expected with a better-designed filter in which preferential flows are minimised.

7 Future research

Based on the knowledge and experience gained through this PhD project, the following ideas and suggestions for future research are provided.

Landfill gas migration and mitigation strategies:

- Carbon dioxide surface screenings, in combination with CH_4 screenings, to evaluate if CH_4 oxidation in the cover leads to enhanced CO_2 emissions and to assess the basic respiration of the landfill cover, to understand better non-loaded CO_2 emissions.
- Further investigations into the lateral migration of landfill gas, to understand better if it is a fair assumption that this migration pathway is insignificant compared to the other pathways.
- Further studies that evaluate the available landfill gas generation models to help understand which model would be the most suitable for a given type of landfill.

Landfill gas screening tool:

- Further research into cost-efficient screening tools for delineating landfill gas emission hotspots is needed. CH_4-specific sensors could potentially overcome some of the limitations of using a TIR camera for emission hotspot screening. Similar tests should be conducted with a CH_4-specific sensor to determine if the delineation of LFG emission hotspots with a limited spatial distribution could be achieved, and if significant emission hotspots and leakages in general could be localised.

Mitigation of diluted LFG:

- A pilot-scale or full-scale biofilter with limited preferential flows should be designed (maybe built into the cover of a landfill) and a new set of tests should be conducted to see if it is possible to find the same high oxidation rates in the field as in the controlled laboratory tests, when preferential flows are limited. Furthermore, the maximum CH_4 oxidation capacity of the filter should be tested when preferential flows are limited.
- Further tests with other CH_4 inlet concentrations and/or higher flow rates should be conducted, including tests with lower CH_4 concentrations similar to what was found, for example, from the remediation systems, in order to prevent lateral migration (concentrations up to 2 vol.%) and higher pumping flows (>10 m^3 h^{-1}).

- Tests to determine a minimum retention time should be conducted, including a test on how retention time and inlet CH_4 concentrations are related. A lower CH_4/O_2 ratio could potentially reduce this time and this would be achieved when LFG is diluted with air to a lower CH_4 concentration or by injecting air at different depths of the filter material. Multiple air injections, to the best of my knowledge, have only been tested in laboratory column tests, and so further research in pilot-scale at a landfill would be needed.
- A biofilter with multiple gas injections (not only air at multiple levels, but diluted LFG) should be tested to determine the performance and efficiency of this design.
- Testing the long-term performance of a field-scale biofilter for diluted LFG, to determine if efficiency is stable over time and throughout all seasons.

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10 Papers

I Fjelsted, L., Christensen, A.G., Larsen, J.E., Kjeldsen, P., Scheutz, C., 2018. Assessment of a landfill methane emission screening method using an unmanned aerial vehicle mounted thermal infrared camera – A field study. *Waste Management* - in press.

II Fjelsted, L., Christensen, A.G., Larsen, J.E., Kjeldsen, P., Scheutz, C., 2018. Closing the methane mass balance for an old closed Danish landfill. Accepted (with revision) by *Waste Management*.

III Fjelsted, L., Scheutz, C., Christensen, A.G., Larsen, J.E., Kjeldsen, P., 2018. Biofiltration of diluted landfill gas in an active loaded open bed compost filter. Submitted to *Waste Management*.

In this online version of the thesis, papers **I-III** are not included but can be obtained from electronic article databases, e.g. via www.orbit.dtu.dk, or on request from DTU Environment, Technical University of Denmark, Bygningstorvet, Building 115, 2800 Kgs. Lyngby, Denmark, info@env.dtu.dk

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